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Impact of adjacent land use on coastal wetland sediments

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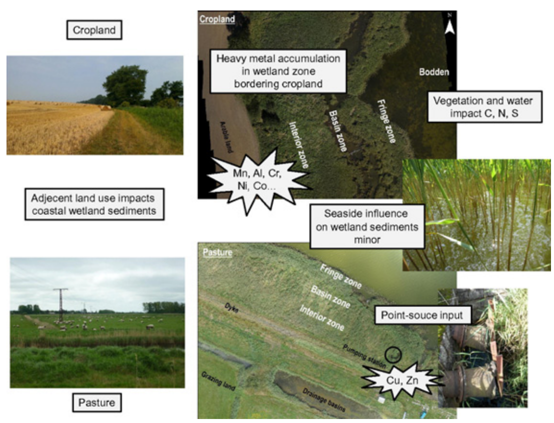
HIGHLIGHTS

- The adjacent land use (cropland/pasture) impacts wetland sediment composition.
- Fertilizer application led to heavy metal accumulation in the zone bordering cropland.
- Seaside influences on sediments were minor compared to influences from land.

GRAPHICAL ABSTRACT



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ABSTRACT

Coastal wetlands link terrestrial with marine ecosystems and are influenced from both land and sea. Therefore, they are ecotones with strong biogeochemical gradients. We analyzed sediment characteristics including macronutrients (C, N, P, K, Mg, Ca, S) and heavy metals (Mn, Fe, Cu, Zn, Al, Co, Cr, Ni) of two coastal wetlands dominated by *Phragmites australis* at the Darss-Zingst Bodden Chain, a lagoon system at the Southern Baltic Sea, to identify the impact of adjacent land use and to distinguish between influences from land or sea. In the wetland directly adjacent to cropland (study site Dabitz) heavy metal concentrations were significantly elevated. Fertilizer application led to heavy metal accumulation in the sediments of the adjacent wetland zones. In contrast, at the other study site (Michaelsdorf), where the hinterland has been used as pasture, heavy metal concentrations were low. While the amount of macronutrients was also influenced by vegetation characteristics (e.g. carbon) or water chemistry (e.g. sulfate), the accumulation of heavy metals is regarded as purely anthropogenic influence. A principal component analysis (PCA) based on the sediment data showed that the wetland fringes of the two study sites are not distinguishable, neither in their macronutrient status nor in their concentrations of heavy metals, whereas the interior zones exhibit large differences in terms of heavy metal concentrations. This suggests that seaside influences are minor compared to influences from land. Altogether, heavy metal concentrations were still below national precautionary and action values. However, if we regard the macronutrient and heavy metal concentrations in the wetland fringes as the natural background values, an accumulation of trace elements from agricultural production in the hinterland is apparent. Thus, coastal wetlands bordering croplands may function as

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effective pollutant buffers today, but the future development has to be monitored closely to avoid breakthroughs due to exceeded carrying capacities.

1. Introduction

Coastal wetlands are open structures with strong interactions along the land-water interface, linking terrestrial with marine ecosystems (Andreu et al., 2016), and represent ecotones in the core sense of the term. Coastal wetlands can provide a variety of ecosystem services that are fundamental for physical processes and biogeochemical cycling including sediment retention and protection against coastal erosion, habitats for fish or birds, raw material provisioning, pollutant buffering and nutrient regulation (Duarte et al., 2013; Karstens and Lukas, 2014; Perillo et al., 2009; Reddy et al., 1999). The relative importance of these services for humans often depends on management decisions and the specific location of a coastal wetland.

Coastal wetland sediments are a mixture of material from various sources including terrestrial input via surface or groundwater flows, erosion of near-by coastal sites or adjacent land, atmospheric depositions or sedimentation of marine particles (Abi-Ghanem et al., 2009; Bao et al., 2015). Macronutrients may have natural origins (wetland vegetation or sea influence), whereas the accumulation of heavy metals in sediments can be regarded as a 'finger print of human activity' (Andreu et al., 2016). Heavy metal concentrations in the landward zones of wetlands may be largely driven by input from adjacent parts of land, especially when these are croplands because erosion tends to be stronger compared to permanent grasslands or forests (Pimentel and Kounang, 1998). In aquatic environments, heavy metals can be a major threat due to their persistence, prevalence, potential toxicity and bioavailability (Boyd, 2010; Marchand et al., 2011). Identifying the sources of heavy metals and evaluating of the influence of anthropogenic activities is difficult (Bayen, 2012; Wang et al., 2014). Agriculture is often mentioned as an important input source and fertilizers applied in agroecosystems can be a major source of heavy metals (Jiao et al., 2012). Some heavy metals included in fertilizers are essential for plant growth but toxic above critical concentrations (e.g. copper, zinc, manganese, iron), whereas others are always contaminants with no benefits for plant growth (e.g. chromium) (He et al., 2005). Contamination assessments and monitoring of heavy metal concentrations in coastal wetland sediments are essential, especially when anthropogenic pressures are on the rise (Andreu et al., 2016; Pascual-Aguilar et al., 2015), and coastal wetlands are 'the last line of defense' before pollutants reach adjacent waters.

In this study we analyze the sediment composition of different coastal wetlands including all macronutrients (C, N, P, K, Mg, Ca, S) as well as eight heavy metals (Mn, Fe, Cu, Zn, Al, Co, Cr, Ni) to answer the question, whether sediment characteristics of coastal wetlands dominated by *Phragmites australis* differ depending on adjacent land use. Two typical and representative sites with respect to land use, topography and hydraulic conditions in the Southern Baltic Sea region were chosen. The wetlands were further subdivided into three zones in order to distinguish between influences from land or sea. We address how adjacent land use (pasture vs. cropland) impacts sediment composition and how influences from the land or sea dominate across the wetland zones. By doing this, we aim to differentiate between 'natural' and 'anthropogenic' influences. While the amount of macronutrients in sediments might be either influenced by vegetation characteristics (e.g. carbon) or water chemistry (e.g. sulfur), the accumulation of heavy metals in sediments is regarded as anthropogenic influence.

2. Research approach and methods

2.1. Study sites

The Darss-Zingst Bodden Chain is a lagoon system with four sub-basins at the Southern Baltic Sea in Germany (Fig. 1). It is a shallow water body with a mean water depth of 2 m. The only connection to the open Baltic Sea is a narrow outlet called Gellenstrom in the northeast (Schumann et al., 2006). Tides do not exist and water exchange with the Baltic Sea is induced meteorologically with inflow situations under strong and persistent northeasterly winds (Selig et al., 2007).

The southern hinterland of the Darss-Zingst Bodden Chain is predominantly used for agriculture (Fig. 1). Lowland areas are dyked and used as grassland, whereas areas with a more pronounced topography are usually not dyked and used as cropland. Consequently two different types of coastal wetlands can be differentiated: coastal wetlands bordering arable fields and coastal wetlands confined by a dyke landwards with pastures in the hinterland. At the coasts of the Darss-Zingst Bodden chain both wetland types are dominated by *Pahragmites australis* (Cav) Trin. Ex Streudel (common reed).

In this study, one site of each type was investigated: the *Phragmites* wetland at Dabitz borders directly cropland, whereas the wetland at Michaelsdorf is 'squeezed' behind a dyke and the hinterland used as pasture for sheep (Fig. 1). The distance between the study sites is about 15 km and consequently climatic conditions do not differ. Both sites are situated at the southern coast of the Bodden system. The sediment textures of the adjacent Bodden sediments are fine to medium sands (Bitschofsky et al., 2015). A detailed description of vegetation, water and sediment characteristics of the wetlands follows in the Results Section 3.1.

Tidal salt marshes are often divided into low, mid- and high marsh according to the influence of the tidal range (Packham and Willis, 1997). Since there are no tides in the Darss-Zingst Bodden Chain the wetlands are subdivided into interior, basin and fringe zone, based on water level and hydraulic energy (Fig. 1). This classification was already proposed in the 1970s for mangrove forests (Lugo and Snedaker, 1974), and proved to be functional. Water level and hydraulic energy are higher in the fringe than in the basin zone, whereas the interior zone is rarely flooded (see Brinson, 1993; Karstens et al., 2015a; Lugo et al., 1988).

2.2. Analysis of land use

Aerial images (1953–2013) provided by the government office for geoinformation, surveying and cadaster MV were used to analyze land use changes at the two study sites since the 1950s. Semi-structured, face-to-face interviews were conducted with the farming company that manages the cropland at Dabitz and with the shepherds responsible for sheep grazing at Michaelsdorf to improve our understanding of past and present land use activities. Questions were grouped into categories, but the order of questions was not pre-defined, and further questions could be added during the interviews to enhance flexibility and allow in-depth information (Hollway and Jefferson, 2000; Helfferich, 2009).

2.3. Sampling and analysis of sediment, water and vegetation

A total of 60 sediment samples at Dabitz and 48 samples at Michaelsdorf were collected between March 2014 and January

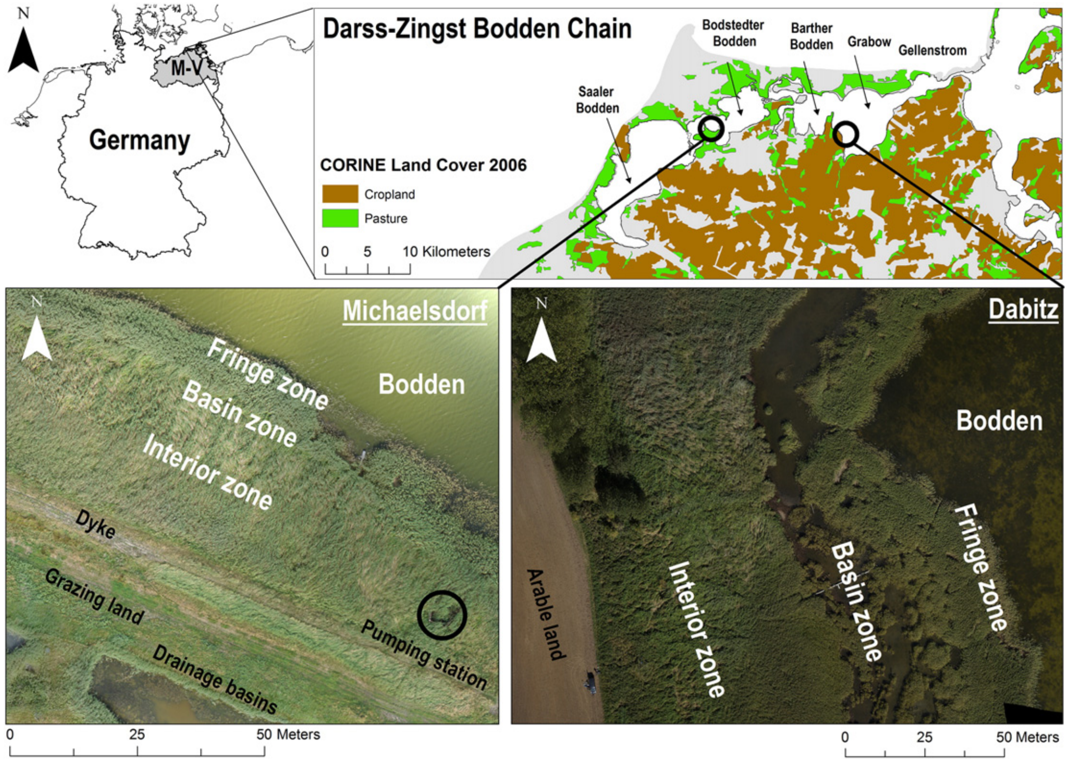

Fig. 1. Study sites Michaelsdorf and Dabitz at the southern coast of the Darss-Zingst Bodden Chain. Areas used as pastures or croplands were extracted from the CORINE Land Cover Dataset of 2006. Aerial images of the study sites were taken in August 2015 with an unmanned aerial system flight.

2015 in the three wetland zones using a stainless steel corer with 7 cm diameter (*Hydrobios*, *Kiel*, *Germany*). Sediment samples were separated into depth slices of 0–2 cm and 2–10 cm. We chose this subdivision, because the upper 2 cm best reflect influences of modern land use, while the lower sediment layer of 2–10 cm reflects the last decades. Sedimentation rates outside the wetlands range between 0.9–1.8 mm per year at the Grabow (Meyer et al., submitted for publication) and 0.2–0.6 mm per year at the Barther Bodden (Müller, 2002). A subset of sediment samples was dried at 105 °C for 24 h, sieved to <2 mm and subsequently grinded to prepare for further analysis. Sediment organic matter was determined gravimetrically by loss-on-ignition (LOI) in a muffle furnace at 550 °C for 4 h. Total carbon, nitrogen and sulfur contents were quantified by combustion in a CNS Analyzer (*Vario Max*, *Elementar*, *Germany*). A multi element analysis including total phosphorus, potassium, calcium, magnesium, iron, aluminum, manganese, zinc, copper, chromium, cobalt and nickel was carried out by induced coupled plasma-mass spectrometry after aqua regia digestion (*Optima 5300 DV*, *Perkin-Elmer*, *USA*) (Wuenscher et al., 2015).

Vegetation characteristics including stem density and aboveground biomass were recorded bimonthly between March 2014 and January 2015. Biomass sampling took place in 20 × 20 cm squares in each zone. In addition, in January 2015 we collected in each wetland zone litter samples by harvesting all on-ground litter in three 20 × 20 cm squares. Aboveground plant and litter material was dried at 60 °C for 48 h to weigh dry biomass (Schieferstein, 1997). Total carbon and nitrogen concentrations of the litter samples were quantified by combustion in a CNS Analyzer (*Vario Max*, *Elementar*, *Germany*). Total phosphorus concentrations were analyzed after aqua regia digestion in an ICP-MS (*Optima 5300 DV*, *Perkin-Elmer*, *USA*).

A total of 17 water samples at Dabitz and 10 water samples at Michaelsdorf were taken in the fringe zones between November 2013 and November 2014. Sulfate was measured photometrically after precipitation with barium. Salinity was measured in situ above the sediment bed using *Hach Lange* sensors. Suspended sediment concentrations were determined by filtration over pre-weighted GF/F filter (e.g. Deborde et al., 2007).

2.4. Data processing

All data analysis was performed and all figures were prepared using the open-source statistical software R (version 3.2.1). Pearson correlation coefficients were calculated for all sediment parameters. Beforehand, scatter plot matrices were used to identify visually if other correlations than linear correlations occur. Principal component analysis (PCA) was applied to investigate if we can characterize groups of individual samples. Correlated sediment variables were combined into factors and sources related to the factors were then identified (e.g. Bai et al., 2011; Bao et al., 2015). The R package FactoMineR was used for the PCA (http://factominer.free.fr/index.html).

3. Results

3.1. Wetland characteristics and land use

The study site Dabitz adjoins fields that have been used as cropland at least since the 1950s: already the first available aerial image from 1953 shows that arable fields directly border the *Phragmites* wetland. The farming company that currently cultivates the field confirmed that arable farming was practiced since about 1945. Nowadays a crop rotation system including oil seed rape, wheat and barley is applied. Harvest takes place during summer and tillage in early autumn. The field lays fallow after harvest for 6–8 weeks. Aerial images show that this usually occurs around August. Herbicides and fungicides are applied twice a year, in autumn and spring. Fertilizers are only applied in spring between March and June depending on the crop. Since 2000 mineral phosphorus fertilizers are no longer applied, instead cow manure is used to cover the phosphorus and potassium demands of plants. Since the amount of applied manure does not satisfy the crop demand for nitrogen, mineral nitrogen fertilizers are applied additionally. Due to the relatively high elevation of the field and the sloping relief towards the coast, the field is not affected by flooding. During heavy rain events, erosion rills may form, giving evidence of periodic solid matter transport into the wetland. The soil type of the cropland is loamy sand. Soil pH varied with topography and was lowest with 6.7 in the sinks adjacent to the coastal wetland. Liming is done every 4–5 years.

The topography at the study site Michaelsdorf differs significantly from Dabitz and does not allow arable land use. The land is flat and the ground water gradient goes from sea to land (see Kliesch et al., in press). Dyke construction took place in the 1970. The hinterland is intensively drained and used for sheep grazing since 1990. Besides sheep manure no additional fertilizers or other agricultural products are applied. The sheep graze between April and October and the herd of sheep rotates once an area is grazed (around 15 sheep per hectare). During the other six months of the year grazing is not allowed due to nature protection restrictions and the area is not used agriculturally during that time of the year.

Although both study sites are situated at the southern site of the lagoon system, they are influenced by water masses of two different sub-basins (Fig. 1). Suspended sediment concentrations were comparable at both fringe sites (Michaelsdorf: 65 ± 51 mg/L; Dabitz: 64 ± 55 mg/L). Due to the higher influence of the Baltic Sea at the outermost Bodden salinity and sulfate concentrations were higher at Dabitz with 10.8 ± 1.5 PSU and 497 ± 134 mg/L sulfate compared to 8 ± 1.4 PSU and 266 ± 141 mg/L at Michaelsdorf.

At both wetlands, the vegetation is dominated by *P. australis*. Biomass, stem density and litter mass were comparable between the interior and basin zones of the two study sites (Table 1). Maximum aboveground biomass occurred in late summer and was 8.2 kg/m^2 in the interior zone of Michaelsdorf and 8.4 kg/m^2 at Dabitz. For the fringe zones of the two study sites vegetation characteristics differed. In the fringe zone at Michaelsdorf stem density was significantly lower than at Dabitz and aboveground biomass remained below 2 kg/m^2 during the vegetation maximum. Almost no *Phragmites* stems survived the winter season and when litter sampling took place in January 2015, no litter was left on the sediment (Table 1).

3.2. Sediment properties in different wetland zones

The characteristics of collected sediment samples are summarized in Table 2. Sediment texture was comparable at the wetland interior and basin zone of both study sites with sandy loams and became coarser towards the fringe zones with fine to medium sands. The highest carbon and nitrogen concentrations occurred in the surface sediments of the interior zone at Michaelsdorf, where also litter mass was highest (Table 1). Sulfur concentrations were highest in the lower sediments of the basin zone at Dabitz. The other macronutrient concentrations were always higher in the surface sediments of the interior or basin zone at Dabitz compared to the fringe zones of the two study sites (Fig. 2, Table 2).

Mean heavy metal concentrations were highest in the interior wetland zone at Dabitz compared to all other zones of the two study sites (Table 2; except iron which is slightly higher in the surface sediments of the basin zone). The differences between the interior zones at Dabitz and Michaelsdorf were significant for all heavy metals except copper and zinc (Fig. 3).

The first two principal components of the PCA explain 78.8% (0–2 cm) and 87.4% (2–10 cm) of the total variance of the element concentrations (Fig. 4). The first principal component covers mainly trace elements such as nickel, chromium or copper which also occur in agricultural fertilizers and indicate an 'anthropogenic' influence. The second principal component is associated with carbon, nitrogen and sulfur and reflects the 'natural' influence from vegetation and water characteristics (see Discussion 4.1 and 4.2). The PCA for the surface sediments (0–2 cm) allows us to divide the factorial plane into four parts: (1) Dabitz interior zone: rich in heavy metals, but comparably poor in carbon, nitrogen and sulfur, (2) Michaelsdorf interior zone: rich in carbon, nitrogen and sulfur, but low heavy metal concentrations, (3) Dabitz basin zone: rich in macronutrients and some heavy metals, and (4) Michaelsdorf basin zone, Michaelsdorf fringe zone and Dabitz fringe zone: low heavy metal as well as low macronutrient concentrations (Fig. 4A, Table 5 Appendix A). For the lower sediment layer (2–10 cm) the interior zone and the fringe zone at Michaelsdorf are not separated in the PCA (Fig. 4B).

None of the analyzed elements at either study site are significantly negatively correlated (Fig. 5). At Dabitz, all heavy metals are significantly positively correlated with each other. This pattern is less pronounced at Michaelsdorf.

4. Discussion

4.1. 'Natural' influences: impact of vegetation and water on sediment composition

The PCA showed that the influence from the seaside is minor compared to the influence from the land: the wetland fringes are not distinguishable with respect to their sediment composition. The only exception was an outlier at Michaelsdorf with 7.9% sulfur. In this sample also the iron concentrations were – with 68 862 mg/kg – tenfold higher than at the other sites. Iron and sulfur were significantly positively correlated at Michaelsdorf (Fig. 5, Table 4 Appendix A) and this outlier might have been driven by an iron-sulfide-compound.

Table 1
Mean aboveground biomass and stem density throughout the year ± standard deviation (n = 21). Litter mass and CNP concentrations in January 2015 (n = 9 for Dabitz and n = 6 for Michaelsdorf).

	Dabitz			Michaelsdorf		
	Interior zone	Basin zone	Fringe zone	Interior zone	Basin zone	Fringe zone
Biomass [kg/m^2]	3.9 ± 3.2	3.4 ± 2.8	2.1 ± 1.3	3.8 ± 2.9	2.6 ± 2.4	0.7 ± 0.7
Stem density [stems/m^2]	375 ± 154	407 ± 119	353 ± 87	408 ± 200	422 ± 151	184 ± 82
Litter mass [g/m^2]	853 ± 23	553 ± 135	394 ± 233	1080 ± 104	353 ± 171	0
Carbon in litter [%]	42.4 ± 0.3	43.3 ± 0.5	38.8 ± 2	44.6 ± 0.2	44.8 ± 0.2	–
Nitrogen in litter [%]	1.6 ± 0.2	1.2 ± 0.3	1.3 ± 0.1	1.9 ± 0.1	1.3 ± 0.1	–
Phosphorus in litter [%]	0.011 ± 0.001	0.009 ± 0.004	0.008 ± 0.001	0.013 ± 0.001	0.005 ± 0.001	–

Table 2
Mean dry bulk density, organic matter (LOI: loss-on-ignition) and element concentrations ± standard deviations (n = 128).

	Dabitz					Michaelsdorf						
	Interior 0–2 cm	Interior 2–10 cm	Basin 0–2 cm	Basin 2–10 cm	Fringe 0–2 cm	Fringe 2–10 cm	Interior 0–2 cm	Interior 2–10 cm	Basin 0–2 cm	Basin 2–10 cm	Fringe 0–2 cm	Fringe 2–10 cm
Density [g/cm^3]	0.6 ± 0.4	0.9 ± 0.2	0.2 ± 0.1	0.3 ± 0.2	0.6 ± 0.5	1.1 ± 0.3	0.1 ± 0.04	0.5 ± 0.1	0.4 ± 0.2	0.6 ± 0.1	0.8 ± 0.5	0.8 ± 0.5
LOI [%]	21.1 ± 14.12	12 ± 7.1	48.3 ± 13.3	36.8 ± 14.4	9.9 ± 7.4	1.3 ± 0.8	65.5 ± 10.8	8.3 ± 4	23.1 ± 4.9	22.1 ± 2.5	4.8 ± 4.6	9.2 ± 16.7
C [%]	11 ± 6.6	5.9 ± 3.1	24.3 ± 6.4	18.9 ± 7	4.9 ± 3.7	0.5 ± 0.3	31.1 ± 7.3	5 ± 3.2	11.7 ± 2.6	10.7 ± 3.1	2.2 ± 2.1	3.8 ± 5.6
N [%]	1 ± 0.5	0.6 ± 0.3	1.8 ± 0.4	1.4 ± 0.5	0.5 ± 0.3	0.1 ± 0.04	2.5 ± 0.5	0.4 ± 0.3	1 ± 0.2	0.9 ± 0.2	0.3 ± 0.2	0.5 ± 0.5
S [%]	0.2 ± 0.1	0.1 ± 0.1	0.8 ± 0.1	1.2 ± 0.7	0.6 ± 0.4	0.1 ± 0.04	0.7 ± 0.11	0.2 ± 0.1	0.3 ± 0.1	0.3 ± 0.1	0.4 ± 0.6	0.3 ± 0.5
P [mg/kg]	1309 ± 548	1187 ± 568	1975 ± 710	794 ± 222	435 ± 165	201 ± 35	1422 ± 245	274 ± 119	765 ± 154	480 ± 114	151 ± 49	118 ± 71
K [mg/kg]	3191 ± 1323	3216 ± 1716	4262 ± 974	3849 ± 1216	1519 ± 845	360 ± 110	1331 ± 260	559 ± 132	1767 ± 380	1893 ± 423	412 ± 216	422 ± 279
Ca [mg/kg]	5963 ± 2623	4489 ± 1753	7194 ± 1895	4817 ± 1280	3732 ± 2357	1419 ± 518	7225 ± 1720	1345 ± 554	5104 ± 1032	3376 ± 589	3136 ± 2607	2507 ± 2885
Mg [mg/kg]	3717 ± 1844	3569 ± 2129	5992 ± 841	5184 ± 1573	2436 ± 1316	603 ± 137	3725 ± 806	735 ± 245	2717 ± 564	2245 ± 549	796 ± 711	1088 ± 1370
Mn [mg/kg]	1062 ± 685	631 ± 361	406 ± 388	118 ± 51	95 ± 46	29 ± 8	36 ± 10	11 ± 3	140 ± 88	33 ± 7	39 ± 34	24 ± 14
Fe [mg/kg]	18,465 ± 8319	21,746 ± 12,024	26,917 ± 17,640	12,725 ± 5131	6831 ± 3646	1846 ± 511	6849 ± 1279	1601 ± 383	17,552 ± 8711	5727 ± 1153	4942 ± 7175	3077 ± 4102
Cu [mg/kg]	18 ± 7	16 ± 6	16 ± 2	18 ± 5	11 ± 4	5 ± 1	16 ± 3	7 ± 2	10 ± 2	9 ± 2	4 ± 2	4 ± 1
Zn [mg/kg]	97 ± 48	74 ± 38	70 ± 18	55 ± 17	45 ± 18	20 ± 8	69 ± 20	17 ± 11	46 ± 21	35 ± 16	15 ± 9	16 ± 9
Al [mg/kg]	11,368 ± 4227	14,151 ± 7817	6302 ± 540	8123 ± 2522	3208 ± 1485	1159 ± 349	2226 ± 714	1596 ± 323	4775 ± 1139	5870 ± 1339	1226 ± 533	1492 ± 786
Co [mg/kg]	7 ± 3.1	6.8 ± 3.1	4.5 ± 2.3	3.5 ± 1.1	2.3 ± 1	0.9 ± 0.3	1.2 ± 0.2	0.6 ± 0.1	2.1 ± 0.5	1.4 ± 0.3	1.4 ± 1.3	1.4 ± 1.4
Cr [mg/kg]	17.7 ± 5.9	20.4 ± 9.5	13.2 ± 1.3	16.6 ± 5.1	7.2 ± 3.5	2.4 ± 0.6	5.6 ± 1.7	3.9 ± 1	10.4 ± 2.4	11.7 ± 2.5	2.4 ± 0.9	2.6 ± 1.2
Ni [mg/kg]	13.1 ± 5.6	12.7 ± 6.2	12.6 ± 1.6	12.4 ± 3.7	7.9 ± 5	2.4 ± 0.8	5.3 ± 0.9	2.3 ± 0.8	7.2 ± 1.2	6.3 ± 1.1	2.8 ± 2.3	3 ± 3.6

Besides this exception, sulfur concentrations were highest in the basin zone at Dabitz. Dabitz is situated relatively near to the Bodden outlet, and sulfate ions are supplied by saltwater inflow from the Baltic Sea through the Gellenstrom (Fig. 1). Under anoxic conditions sulfate is reduced and can react with iron compounds and precipitate as FeS (e.g. Boström et al., 1988). Sediments in the fringe zone at Dabitz are generally well oxygenated due to higher turbulence and also the surface layer in the basin zone becomes anoxic only occasionally (Karstens et al., 2015a). The lower sulfur contents in the upper two centimeters in the basin zone and in the fringe zone compared to the second layer suggest a decreased efficiency of sulfate reduction in the lower sediments.

In the interior zone at Michaelsdorf, we measured the highest carbon, nitrogen and phosphorus stocks in the litter mass compared to all other zones of both study sites. This zone is well protected and rarely flooded. Therefore, litter can be directly incorporated into the sediment. The high carbon, nitrogen and phosphorus concentrations in the surface sediments support this hypothesis. In this zone, *Phragmites* affects the sediment compositions due to its high biomass production and litter abundance and functions therefore as a habitat-modifier (see also Rooth et al., 2003). High biomass production in combination with long residence times of litter material on the sediment floor can increase carbon accumulation rates 5-fold in *Phragmites australis* stands compared to *Spartina patens* stands (Windham, 2001). At Dabitz, aboveground biomass and litter mass were slightly lower in the basin than in the interior zone; however carbon, nitrogen and phosphorus concentrations in the surface sediments were significantly higher in the basin zone (Fig. 2). The basin zone is water-logged most of the year. Shallow inundation, alternating desiccation-flooding-cycles and damp conditions favor decomposition of *Phragmites* litter (Bedford, 2005; Völlm and Tanneberger, 2014). *Phragmites* is a strong source of carbon, nitrogen and phosphorus in both the interior and basin zone, but the hydrological conditions determine the further fate of those inputs. Additionally, the basin zone at Dabitz is an accumulation area for fine-grained particles rich in organic matter and phosphorus supplied from the Bodden (Karstens et al., 2015a). Although vegetation density and litter accumulation were higher in the fringe zone at Dabitz than at Michaelsdorf, the wetland edges differed only slightly and not significantly in their sediment composition (Figs. 2 and 3). Neither vegetation nor water characteristics seem to be sufficiently different at the study sites to produce an overall difference in sediment composition of the wetland fringes (Fig. 4).

4.2. 'Anthropogenic' influences: impact of adjacent land use on sediment composition

At Dabitz, where the wetland directly borders arable land, heavy metal concentrations were significantly higher in the interior and in the basin zones compared to the fringe zone (Fig. 3). Besides the six essential macronutrients (N, P, K, Ca, Mg, S), agricultural fertilizers often contain trace elements (Gupta et al., 2014). Therefore, agricultural production and fertilizers applied in the hinterland are often identified as one major input source of heavy metals into adjacent coastal wetlands (Wang et al., 2014). Among the analyzed heavy metals, iron, manganese, copper, zinc and nickel are essential for plant growth, but can be toxic to plants at high concentrations (He et al., 2005). However, other trace metals found frequently in agroecosystem are not essential for plant growth and are toxic for living organisms, including the analyzed chromium (He et al., 2005). The topography of the hinterland at Dabitz is undulating and the elevation decreases towards the perimeter of the Bodden. Therefore, erosion and particle transport from the arable land towards the adjacent wetland seems likely, especially when the cropland lays fallow for 6–8 weeks during summer or when erosion rills form after heavy rain events.

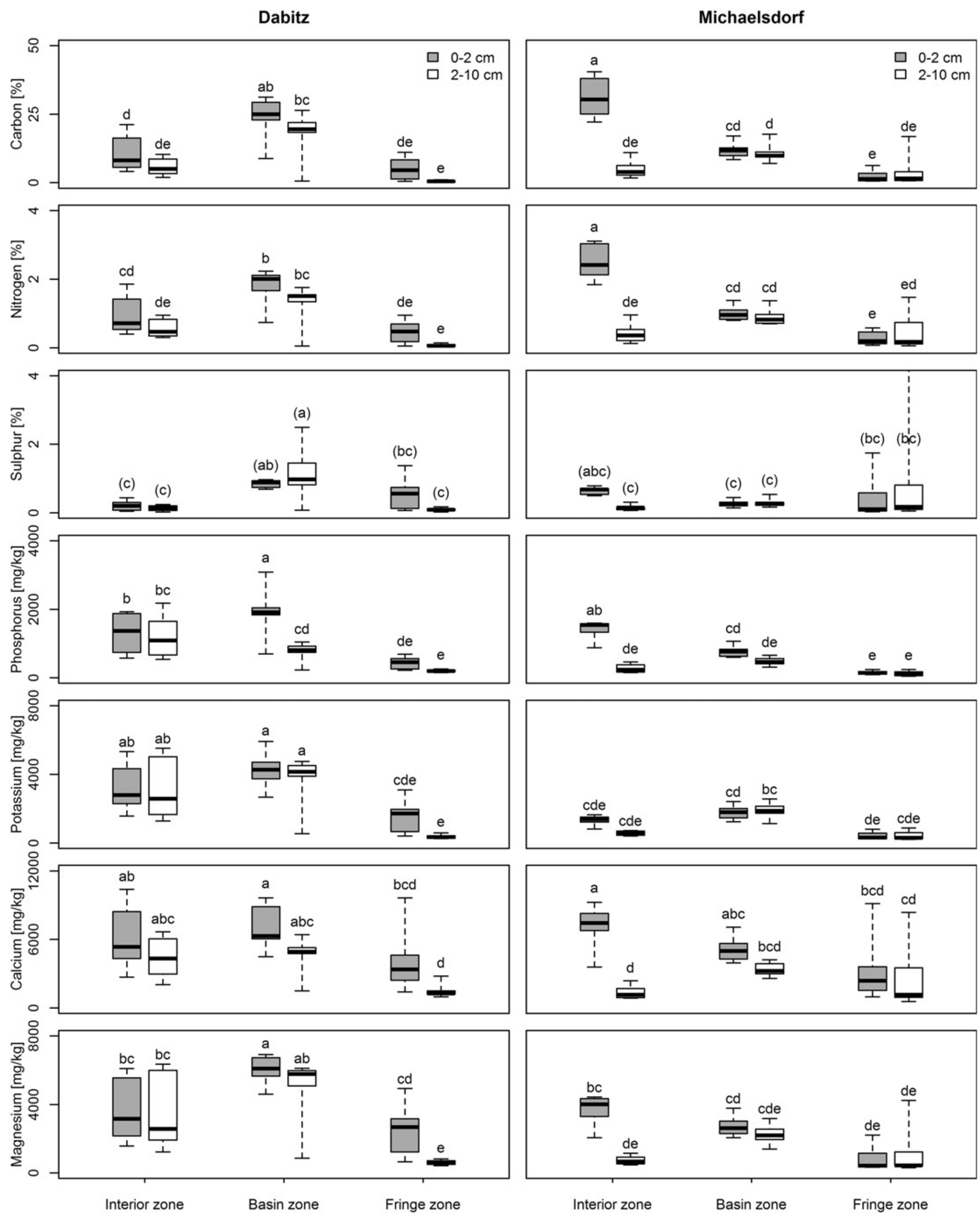

Fig. 2. Macronutrients in coastal wetland sediments: carbon, nitrogen, sulfur in [%], phosphorus, potassium, calcium and magnesium in [mg/kg]. The central mark of each box is the median; the edges display the 25th and 75th percentiles. The whiskers extend to the most extreme data points. Letters represent the results of post-hoc comparisons of group means with Tukey's honest significant differences test ($p < 0.05$) which were conducted between all zones of the two study sites (one outlier in sulfur concentrations was removed for testing).

There were no significant differences in heavy metal concentrations in the interior zone at Dabitz between the surface sediments and the sediment layer below, except for manganese (Fig. 3). This suggests that accumulation of trace elements started already in the past. The hinterland at Dabitz has been used for crop production for more than 60 years and agricultural activities continue to influence the adjacent coastal wetland. Especially phosphorus fertilizers can be a strong source of heavy metal input into agroecosystems (He et al., 2005). Kratz et al. (2015) analyzed trace elements in five different types of mineral phosphorus fertilizers sold in Germany. Most fertilizers contained high amounts of heavy metals, even if not declared as such on the product package (Kratz et al., 2015). Mean element concentrations in straight phosphorus fertilizers were 7289 mg iron, 2934 mg aluminum, 354 mg zinc, 138 mg manganese, 135 mg chromium, 43 mg copper and 24 mg nickel per kg fertilizer (Kratz et al., 2015). Compared to phosphorus fertilizers, nitrogen fertilizers contain only negligible amounts of heavy metals (Kördel et al., 2007). Since 2000, mineral phosphorus fertilizers are no longer applied at the study site Dabitz because the farming company shifted to cow manure. Cow manure contains a tenfold higher amount of zinc and copper and twice as much nickel as mineral phosphorus fertilizers (Kratz and Schnug, 2005). Kratz and Schnug (2005) did not include manganese in their study; however German Chambers of Agriculture report higher manganese contents than zinc in cow manure (e.g. Landwirtschaftskammer NRW, 2015).

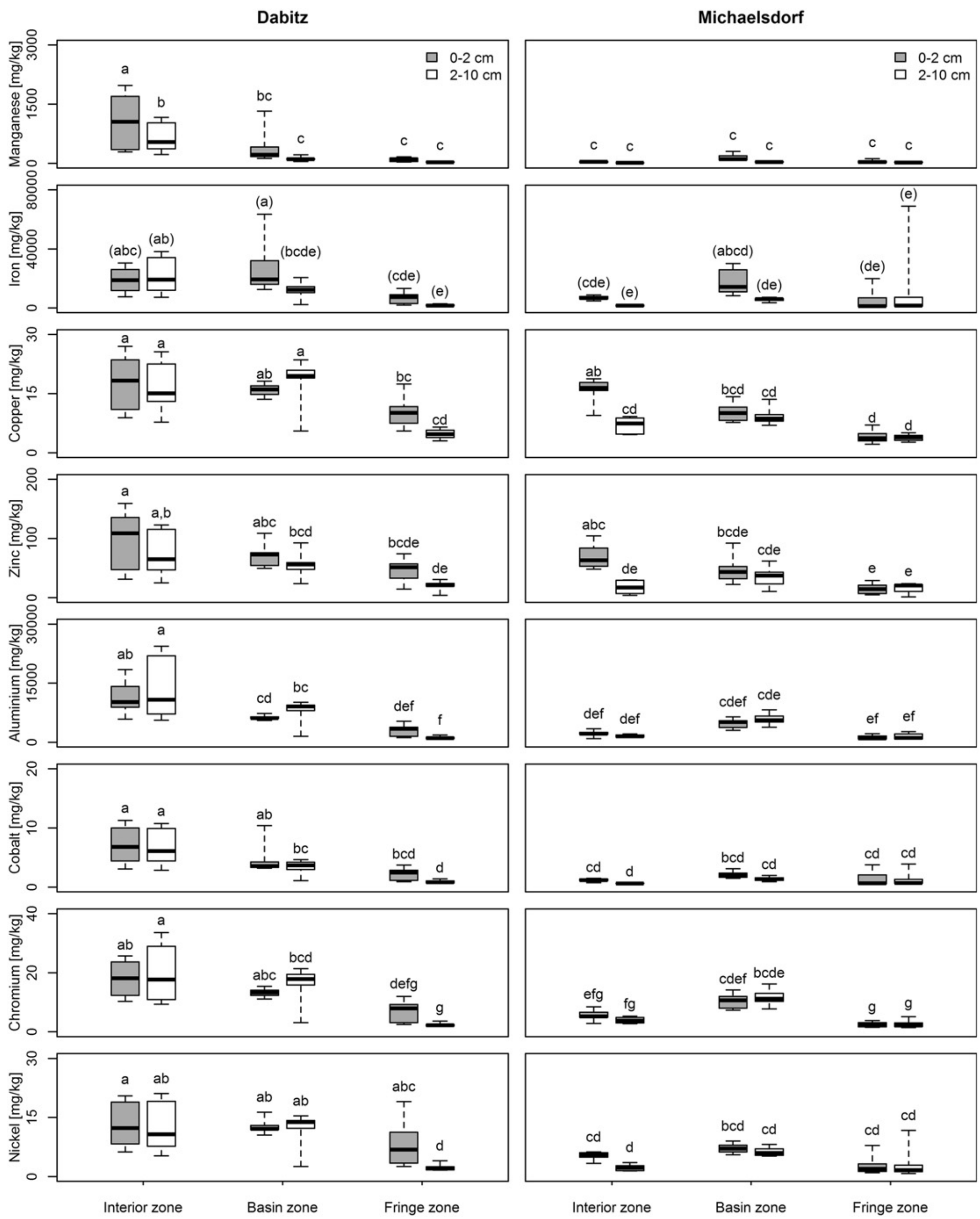

Fig. 3. Heavy metal concentrations [mg/kg]: manganese, iron, copper, zinc, aluminum, cobalt, chromium, nickel. Letters represent the results of post-hoc comparisons of group means with Tukey's honest significant differences test ($p < 0.05$) which were conducted between all zones of the two study sites (one outlier in iron concentrations was removed for testing).

Consequently, the manganese input at Dabitz probably increased since the shift from mineral fertilizers to farm manure. Chromium concentrations are lower in cow manure than in mineral fertilizers (Kratz and Schnug, 2005), thus we expect a decrease in chromium input. In accordance with that, mean manganese, zinc, copper and nickel concentrations were higher in the surface sediments of the interior zone, while chromium concentrations were lower (Fig. 3).

Mean heavy metal concentrations in the fringe zone at Dabitz are significantly lower than in the interior and basin zones, indicating that particle transport from land to sea has been inhibited until now. This is in agreement with tracer test studies showing that dense *Phragmites* stands effectively suppress particle transport in the interior zone (Karstens et al., 2015b). However, redox-sensitive elements that accumulated due to agricultural activities may dissolve in the temporarily anoxic basin zone and be transported in solution into the adjacent Bodden water. The release of phosphorus under anoxic conditions in the basin zone at Dabitz has been discussed by Karstens et al. (2015a). For heavy metals, the mobility depends not only on total concentrations but also on sediment properties such as pH or amounts and forms of oxides (He et al., 2005). Depending on the surrounding environmental conditions and temporal dynamics coastal sediments can switch from sinks to sources for heavy metals (El Nemr et al., 2015; Rai, 2009). In the basin zone at Dabitz, environmental conditions and especially the oxygen status change quickly, and consequently there is a potential risk of heavy metal release into the Bodden water.

At Michaelsdorf heavy metal concentrations in sediments were low with two exceptions: zinc and copper in the surface sediments of the interior zone. One possible explanation could be leaching from the

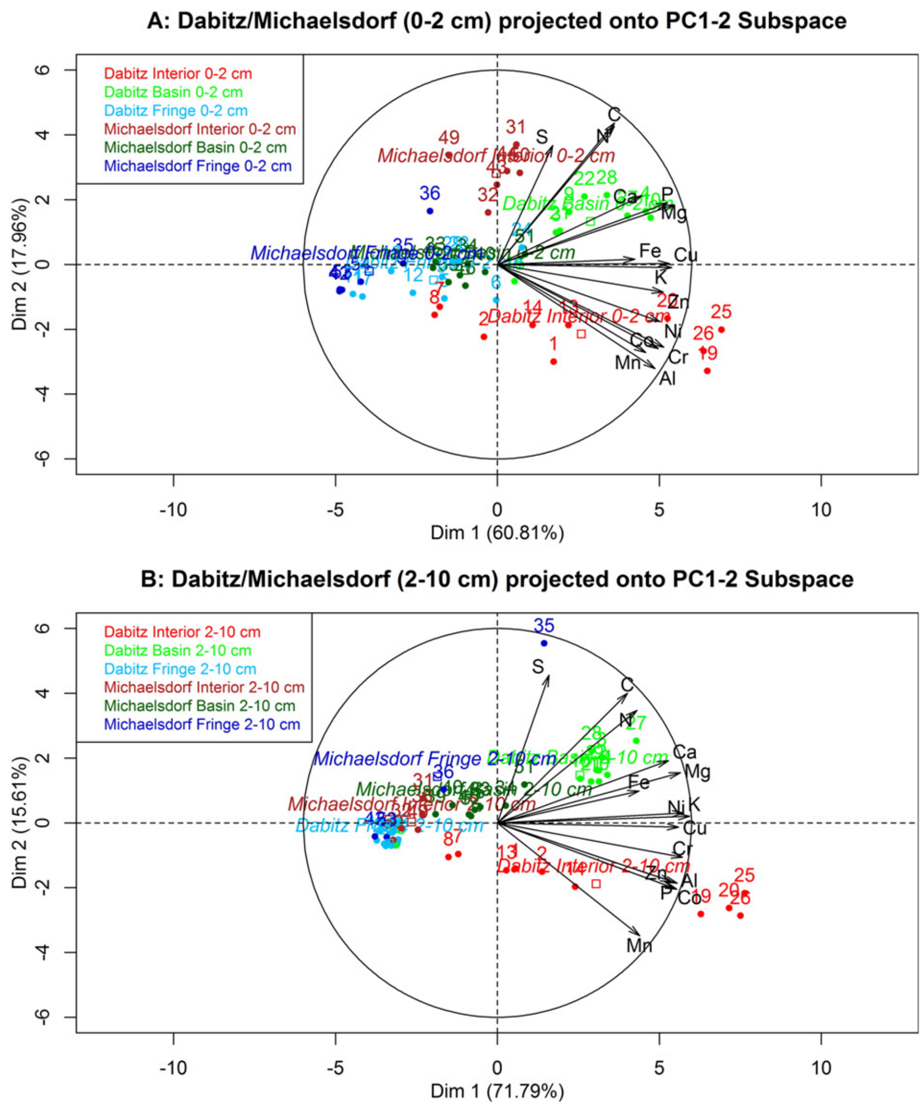

Fig. 4. Principal component analysis for (A) the surface sediments (0–2 cm) and (B) the lower sediment layer (2–10 cm). For correlations between the first two dimensions and sediment variables see Tables 5 and 6 in Appendix A.

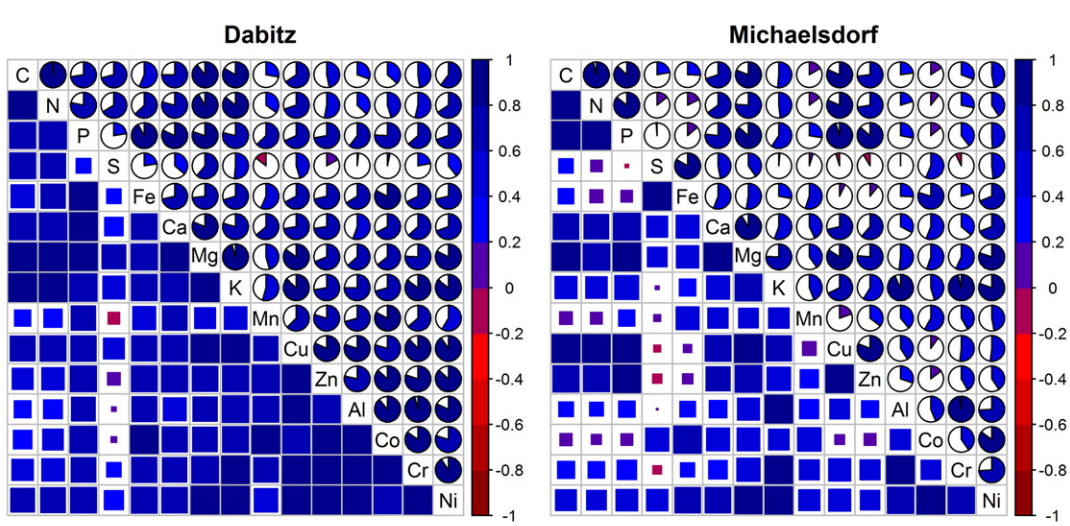

Fig. 5. Graphical display of Pearson correlation matrix of element concentrations at Dabitz and Michaelsdorf. In the lower triangle the biggest squares represent the highest correlations. In the upper triangle a full ellipse stands for a correlation = 1 and an empty ellipse = 0. For correlations numbers and significance levels please refer to Tables 3 and 4 in Appendix A.

rusted construction material of the pumping station (see Fig. 1). The pumping station was installed back in 1970 and the material corrodes. Another possible explanation could be input of zinc and copper from sheep manure. In this case, manure is not applied additionally but left by the grazing sheep. Solid sheep manure contains naturally high amounts of zinc and copper (149 mg zinc and 15 mg copper per kg dry mass; Kördel et al., 2007). However, sheep grazing only takes place between April and October and the herd of sheep rotates. A point-sourced input of copper and zinc from rusting metal seems more realistic. At the moment the pumping station at Michaelsdorf is no longer in operation. However, repeated input of water from channels that drain the hinterland could cause trace element enrichment at a larger scale and contaminations beyond localized point-sources: It has been reported, that field drainage networks may transport pollutants from the hinterland to receiving surface water bodies (He et al., 2005; Hill and Robinson, 2012).

Bao et al. (2015) summarized heavy metal concentrations including copper, zinc, chromium and nickel in coastal wetland sediments at 25 different study sites worldwide. The concentrations of copper, zinc, chromium and nickel at Dabitz do not exceed these values and are within the mid-range of reported concentrations. Only manganese concentrations in the surface sediments of the interior zone at Dabitz are – with average 1062 mg/kg – particularly high and above average values measured at other coastal wetlands (e.g. 733 mg/kg Bao et al., 2015; 1013 mg/kg Fernandes et al., 2011; 392 mg/kg Morillo et al., 2004; 127 mg/kg Wang et al., 2014). Heavy metal concentrations at Dabitz and Michaelsdorf are still below the precautionary and action values of the German Federal Soil Protection Ordinance (BBodSchV, 1999). Also the potential ecological risk level for zinc, copper, nickel and chromium, developed by Hakanson (1980) for coastal sediments, is still low (<40). The risk index ranges from 'low' (<40) over 'moderate' and 'considerable' to 'high' and 'serious' (>320) and is based on the measured heavy metal concentrations, pre-industrial background values and a 'toxic-response' factor for the given substance (see also El Nemr et al., 2015 or Li et al., 2015 for ecological risk index method). However, if we assume that the concentrations in the fringe zones can be regarded as the natural background value, an enormous accumulation of heavy metals from agricultural production in the hinterland has to be assumed.

4.3. Management options inside coastal wetlands

Phragmites has been used traditionally in northern Germany for construction material (Köbbing et al., 2013). Also along the coast of the Darss-Zingst Bodden Chain, our aerial image analysis showed large areas where reed biomass was harvested in February or March but reliable and large scale data on the amount of harvest does not exist yet. However, several studies have proposed phytoremediation as a green technology to remove heavy metals from polluted soils (e.g. McIntyre, 2003; Raskin et al., 1997; Rai, 2009; Salt et al., 1995). It is regarded as an environmentally friendly and cost-effective technology, where the success depends mainly on plant characteristics: the vegetation has to produce large biomass in short time-frames, accumulation of heavy metals in shoots has to be high and harvest has to be easy (He et al., 2005). Southichak et al. (2006) proposed *Phragmites australis* as a biosorbent for the removal of heavy metals. *Phragmites* contains high amounts of lignin and cellulose which allow the adsorption of heavy metals even at low concentrations (Srivastava et al., 1994). Furthermore, *Phragmites* is very resistant to polluted environments and grows fast with high biomass production (Southichak et al., 2006). *Phragmites* harvest in northern Germany takes usually place in winter (Köbbing et al., 2013). To be most effective, harvest should take place when biomass and heavy metal content reach maximum values. This occurs for *Phragmites* during early autumn before senescence (Bragato et al., 2006). However, harvest in autumn contradicts with the objectives of nature protection in our study region (e.g. bird protection: arrival of cranes in early autumn). Furthermore, the removal of biomass would influence the capacity of the *Phragmites* wetland to suppress particle transport and could potentially accelerate erosion processes and, thus, transfer of heavy metals towards the fringe zones. Presumably, measures directly applied on the cropland might be more appropriate to prevent a transfer from heavy metals into the adjacent coastal wetlands.

5. Conclusions

This study demonstrated that land use activities adjacent to coastal wetlands can have a large impact on the sediment composition. Heavy metal concentrations are significantly elevated in the wetland zone that borders directly an arable field where crop production with fertilizer application took place at least since the 1950s. In contrast to this, heavy metal concentrations are low in the coastal wetland that is confined by a dyke with sheep grazing in the hinterland. The only exceptions are comparably high zinc and copper concentrations in the surface sediments of the interior zone, possibly resulting from point-source leaching of the rusted pumping station at the study site. Influences from the sea on coastal wetland sediment compositions are minor compared to the influences from land: The two wetland fringes do not differ significantly, neither in their macronutrient status nor in their concentrations of heavy metals. While the anthropogenic activities in the hinterland seem to impact the heavy metal accumulation, 'natural' differences in vegetation and water characteristics in the different wetland zones likely influence carbon, nitrogen, phosphorus and sulfur concentrations. Coastal wetlands provide a variety of ecosystem functions and which of these are seen as services often depends on management decisions, including decisions about land use in the hinterland. At Dabitz, the *Phragmites* wetland had to become an active pollutant buffer. Until now heavy metals are accumulated and retained in the interior and basin zones, but future developments should be monitored closely to avoid breakthroughs due to exceeded carrying capacities. Further, the possibility of heavy metal release into the water in the temporarily anoxic basin zone should be investigated.

Acknowledgments

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Appendix A

Table 3
Correlation matrix for element concentrations at study site Dabitz. Bold number = correlation is significant at the 0.01 level (2-tailed). Gray background = Pearson correlation coefficient is higher than 0.7.

	C	N	P	S	Fe	Ca	Mg	K	Mn	Cu	Zn	Al	Co	Cr	Ni
C	1	0.99	0.73	0.71	0.56	0.75	0.89	0.84	0.28	0.66	0.47	0.3	0.37	0.48	0.6
N	0.99	1	0.78	0.66	0.63	0.79	0.92	0.86	0.35	0.7	0.53	0.37	0.45	0.54	0.65
P	0.73	0.78	1	0.23	0.92	0.82	0.8	0.79	0.63	0.68	0.72	0.6	0.76	0.64	0.7
S	0.71	0.66	0.23	1	0.22	0.36	0.61	0.52	−0.15	0.45	0.17	0.02	0.03	0.22	0.39
Fe	0.56	0.63	0.92	0.22	1	0.71	0.73	0.7	0.57	0.67	0.71	0.66	0.83	0.67	0.72
Ca	0.75	0.79	0.82	0.36	0.71	1	0.81	0.79	0.64	0.72	0.73	0.55	0.67	0.63	0.7
Mg	0.89	0.92	0.8	0.61	0.73	0.81	1	0.96	0.46	0.85	0.69	0.63	0.65	0.76	0.83
K	0.84	0.86	0.79	0.52	0.7	0.79	0.96	1	0.54	0.87	0.72	0.75	0.71	0.85	0.86
Mn	0.28	0.35	0.63	−0.15	0.57	0.64	0.46	0.54	1	0.62	0.79	0.71	0.83	0.64	0.59
Cu	0.66	0.7	0.68	0.45	0.67	0.72	0.85	0.87	0.62	1	0.82	0.8	0.79	0.88	0.9
Zn	0.47	0.53	0.72	0.17	0.71	0.73	0.69	0.72	0.79	0.82	1	0.79	0.87	0.8	0.87
Al	0.3	0.37	0.6	0.02	0.66	0.55	0.63	0.75	0.71	0.8	0.79	1	0.89	0.96	0.82
Co	0.37	0.45	0.76	0.03	0.83	0.67	0.65	0.71	0.83	0.79	0.87	0.89	1	0.85	0.8
Cr	0.48	0.54	0.64	0.22	0.67	0.63	0.76	0.85	0.64	0.88	0.8	0.96	0.85	1	0.91
Ni	0.6	0.65	0.7	0.39	0.72	0.7	0.83	0.86	0.59	0.9	0.87	0.82	0.8	0.91	1

Table 4
Correlation matrix for element concentrations at study site Michaelsdorf. Bold number = correlation is significant at the 0.01 level (2-tailed). Gray background = Pearson correlation coefficient is higher than 0.7.

	C	N	P	S	Fe	Ca	Mg	K	Mn	Cu	Zn	Al	Co	Cr	Ni
C	1	0.96	0.87	0.22	0.25	0.7	0.81	0.51	0.16	0.82	0.73	0.23	0.16	0.31	0.47
N	0.96	1	0.86	0.14	0.17	0.65	0.76	0.48	0.15	0.82	0.72	0.2	0.11	0.28	0.41
P	0.87	0.86	1	−0.01	0.14	0.77	0.86	0.59	0.28	0.95	0.86	0.28	0.14	0.39	0.49
S	0.22	0.14	−0.01	1	0.83	0.47	0.4	0.01	0.05	−0.05	−0.09	0	0.56	−0.07	0.48
Fe	0.25	0.17	0.14	0.83	1	0.56	0.53	0.28	0.55	0.07	0.11	0.26	0.8	0.21	0.69
Ca	0.7	0.65	0.77	0.47	0.56	1	0.91	0.56	0.43	0.71	0.61	0.33	0.56	0.36	0.69
Mg	0.81	0.76	0.86	0.4	0.53	0.91	1	0.75	0.4	0.84	0.77	0.52	0.55	0.57	0.8
K	0.51	0.48	0.59	0.01	0.28	0.56	0.75	1	0.44	0.68	0.59	0.93	0.45	0.96	0.81
Mn	0.16	0.15	0.28	0.05	0.55	0.43	0.4	0.44	1	0.2	0.36	0.39	0.58	0.42	0.47
Cu	0.82	0.82	0.95	−0.05	0.07	0.71	0.84	0.68	0.2	1	0.81	0.41	0.1	0.52	0.52
Zn	0.73	0.72	0.86	−0.09	0.11	0.61	0.77	0.59	0.36	0.81	1	0.3	0.15	0.42	0.4
Al	0.23	0.2	0.28	0	0.26	0.33	0.52	0.93	0.39	0.41	0.3	1	0.44	0.98	0.74
Co	0.16	0.11	0.14	0.56	0.8	0.56	0.55	0.45	0.58	0.1	0.15	0.44	1	0.4	0.85
Cr	0.31	0.28	0.39	−0.07	0.21	0.36	0.57	0.96	0.42	0.52	0.42	0.98	0.4	1	0.74
Ni	0.47	0.41	0.49	0.48	0.69	0.69	0.8	0.81	0.47	0.52	0.4	0.74	0.85	0.74	1

Table 5
Principal component analysis for sediment samples 0–2 cm depth: correlations between first two dimensions and sediment variables. Only variable were significance level is <0.05 are displayed. Results of *dimdesc* function of R package Factominer.

Dimension 1

	Correlation	pvalue
Mg	0.91	1.52E−19
K	0.9	3.04E−18
Cu	0.89	2.95E−17
P	0.87	2.18E−16
Cr	0.86	4.43E−15
Zn	0.85	6.45E−15
Ni	0.84	7.78E−14
Co	0.83	2.28E−13
Al	0.81	1.68E−12
Mn	0.76	1.91E−10
Ca	0.74	1.01E−09
Fe	0.71	1.36E−08
C	0.6	4.57E−06
N	0.6	5.03E−06
S	0.28	0.048
	R^2	pvalue
Zones	0.63	2.01E−08
	Estimate	pvalue
Dabitz basin 0–2 cm	3.13	0.0011
Dabitz interior 0–2 cm	2.84	0.0018
Dabitz fringe 0–2 cm	−1.72	0.0203
Michaelsdorf fringe 0–2 cm	−3.71	0.0004

Dimension 2

	Correlation	pvalue
C	0.73	3.22E−09
N	0.71	1.13E−08
S	0.61	2.95E−06
Ca	0.35	0.013
P	0.32	0.027
Mg	0.31	0.032
Ni	−0.29	0.041
Cr	−0.42	0.002
Co	−0.43	0.002
Mn	−0.45	0.001
Al	−0.54	7.42E−05
	R^2	pvalue
Zones	0.85	1.53E−16
	Estimate	pvalue
Michaelsdorf interior 0–2 cm	2.62	7.68E−07
Dabitz basin 0–2 cm	1.14	0.0063
Dabitz interior 0–2 cm	−2.33	2.30E−07

Table 6
Principal component analysis for sediment samples 2–10 cm depth: correlations between first two dimensions and sediment variables. Results of *dimdesc* function of R package Factominer.

Dimension 1

	Correlation	pvalue
Ni	0.99	2.88E−38
K	0.96	1.10E−25
Cr	0.95	1.78E−24
Mg	0.94	2.16E−22
Cu	0.93	4.74E−21
P	0.92	4.64E−20
Al	0.92	7.19E−20
Co	0.91	5.91E−18
Zn	0.9	2.77E−17
Ca	0.88	8.23E−16
Mn	0.73	7.31E−09
Fe	0.73	9.51E−09
N	0.72	1.97E−08
C	0.67	3.61E−07

	R^2	pvalue
Zones	0.63	1.10E−07

	Estimate	pvalue
Dabitz interior 2–10 cm	3.53	0.0006
Dabitz basin 2–10 cm	3.03	0.0048
Dabitz fringe 2–10 cm	−2.86	0.0004

Dimension 2

	Correlation	pvalue
S	0.76	9.08E−10
C	0.67	4.36E−07
N	0.58	2.52E−05
Ca	0.32	0.03
Zn	−0.3	0.041
P	−0.31	0.036
Co	−0.33	0.023
Al	−0.34	0.02
Mn	−0.58	2.28E−05

	R^2	pvalue
Zones	0.66	2.18E−08

	Estimate	pvalue
Dabitz basin 2–10 cm	1.32	0.0003
Dabitz interior 2–10 cm	−2.03	1.12E−06

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