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**DOI**

[10.1016/j.scitotenv.2020.138233](https://doi.org/10.1016/j.scitotenv.2020.138233)

**Publication date**

2020

**Document Version**

Final published version

**Published in**

Science of the Total Environment

**License**

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[Link to publication](#)

**Citation for published version (APA):**

Pit, I., Wassen, M. J., Kooijman, A. M., Dekker, S. C., Griffioen, J., Arens, S. M., & van Dijk, J. (2020). Can sand nourishment material affect dune vegetation through nutrient addition? *Science of the Total Environment*, 725, [138233]. <https://doi.org/10.1016/j.scitotenv.2020.138233>

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## Can sand nourishment material affect dune vegetation through nutrient addition?



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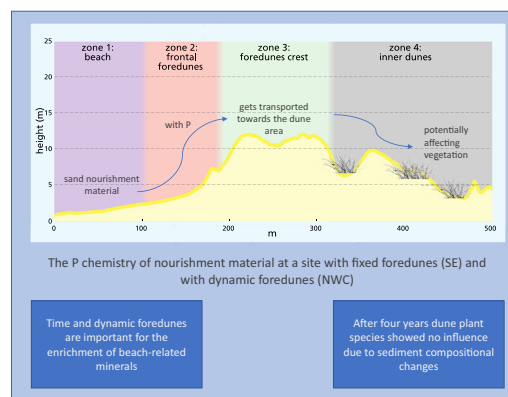
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### HIGHLIGHTS

- Dutch coastlines are commonly nourished with sand from the seabed.
- It is unclear if P chemistry is altered when beach sand enters the dune system.
- Nourishment material was investigated at a site with and without dynamic foredunes.
- After 4 years dune plant species were unaffected by sediment composition changes.
- Enrichment of beach-related minerals depended on time and dynamic foredunes.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 13 November 2019

Received in revised form 24 March 2020

Accepted 25 March 2020

Available online 28 March 2020

Editor: Paulo Pereira

#### Keywords:

Dynamic foredunes

P availability

P speciation

Geochemical characteristics

Beach

Sand engine

### ABSTRACT

In the Netherlands it is common to nourish the coastline with sand from the seabed. Foredunes are replenished with sand from the beach and can be transported further into the dune area. We investigated whether nourishment material alters the phosphorus (P) content of dune soil and the nitrogen (N):P ratio of dune vegetation in two areas: a mega sand nourishment with fixed foredunes (SE) and a traditional sand nourishment with dynamic foredunes (NWC). Four zones were considered: beach (zone 1), frontal foredunes (zone 2), foredunes crest (zone 3) and inner dunes (zone 4). We estimated the characteristics of fine (< 250- $\mu\text{m}$ ) and coarse (250–2000  $\mu\text{m}$ ) sand. Total P, P speciation and available P of SE and NWC were similar until zone 4. Zone 1–3 consisted mainly of coarse sand, whereas the sand in zone 4 was finer with higher amounts at NWC. Iron (Fe) bound P was comparable for fine and coarse sand in zone 1–3, but high contents were present in zone 4. In zone 1–3, calcium (Ca) bound P was mainly found in the fine fraction, which was abundant in the coarse fraction of zone 4. After a period of 4 years, the effect of dynamic dunes on P fractions and dune plant species was not apparent yet, although inblowing sand mainly consisted of fine sand with high contents of Ca-bound P. This may change over time, especially in dynamic dunes with higher eolian activity of fine sand. Consequently, pH buffering of the soil may increase because of a higher Ca-carbonate content, which leads to decreased solubility of Ca-bound P and low P

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availability for the vegetation. Both low P availability and high buffering capacity are known environmental factors that facilitate endangered dune plant species.

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## 1. Introduction

Nutrient availability is the most important factor determining the growth of plants and the productivity of ecosystems in temperate regions (Bedford et al., 1999). In natural, undisturbed ecosystems, nitrogen (N) and phosphorus (P) are the main growth-limiting nutrients (Elser et al., 2007). Widespread N enrichment through human activities is a serious problem for many aquatic and terrestrial ecosystems, leading to algal blooms and anoxic conditions in lakes and rivers and concomitant species extinction (Conley et al., 2009; Rockström et al., 2009). In terrestrial ecosystems, N eutrophication boosts productivity and leads to dominance of fast-growing species that outcompete other plant species, leading to net species loss (Aerts and Bobbink, 1999; Bobbink et al., 2010). N has therefore been the focus in many nutrient studies (e.g. Galloway et al., 2004; Phoenix et al., 2006). Similarly, there is evidence for the detrimental effects of P enrichment for natural ecosystems (Carpenter, 2005; Qualls and Richardson, 2000; Wassen et al., 2005). P and its availability (the portion of P in the soil that can be readily absorbed and assimilated by plants) is essential for plant growth and development (Brady and Weil, 2008; Zheng and Zhang, 2000).

Compared to ecosystems with more fertile soils, dune ecosystems have low to very low soil nutrient availability (e.g. Kellman and Roulet, 1990; Olff et al., 1993). Generally, plant growth in sandy dunes with low organic matter content is limited primarily by N and secondarily by P, both in dry dune systems (Walker and Syers, 1976) and in wet dune slacks (Lammerts and Grootjans, 1997). The relative availability of N and P is well reflected in the N:P ratio of plant material and therefore N:P ratios are regarded as reliable proxies for the relative availability of N and P (Olde Venterink et al., 2003; Verhoeven et al., 1996; Wassen et al., 2005). The availability of P for the uptake by plants depends on the dissolution of minerals in which it is fixed and desorption from sorption sites. The P adsorption complex is influenced by pH and presence of calcium (Ca), iron (Fe), and aluminum (Al), and organic matter (Kooijman et al., 1998; Paludan and Morris, 1999).

Beach and dune sands along the Dutch coast show a transition in CaCO<sub>3</sub> content and become less calcareous northwards (Baak, 1936; Eisma, 1968). The dunes in the southern part are calcareous and show dominance of P fixed to Ca (Lindsay and Schwab, 1982). As a result, N and P co-limit plant growth, but due to acidification by atmospheric deposition or in late successional soils, decalcification enhances dissolution of Ca phosphates and N deposition enhanced the availability of N (Kooijman and Besse, 2002). The availability of P then increases, except when Fe and/or Al oxyhydroxide content of the soil is high enough for P sequestration (Lindsay and Moreno, 1960; Zheng and Zhang, 2000). In the northern part of the Netherlands, dune sands are more acid. For example, in the carbonate- and Fe-poor Wadden district, P is more readily available because there are insufficient Fe and Al oxyhydroxides to form phosphate complexes, resulting in relatively loosely bound P (Kooijman et al., 1998).

Eolian dynamics constantly replenish the dunes, especially the foredunes, with new sand originating from the beach (Arens et al., 2013a). Since 1990 the Dutch coastal system has been artificially supplied with sand: on average, 12 million m<sup>3</sup> of sand is applied along the 350 km coastline yearly (Bakker et al., 2012; Mulder and Tonnon, 2011), and around 25% of this volume is blown into the dunes (Arens et al., 2013b). Sand for these sand nourishments is dredged from the seafloor and deposited on the beach or foreshore. In anticipation of sea level rise and increased frequency and strength of storms, new coastal protection strategies have been designed, one of which is the

mega sand nourishment called the Sand Engine (Stive et al., 2013). The Sand Engine consists of 21 million m<sup>3</sup> of sand supplied in one go in 2011, making it much larger than traditional nourishments of 3–5 million m<sup>3</sup> (Van Dalen and Aarninkhof, 2009).

At the Sand Engine, high amounts of P and Fe oxides were found at the surface, with maximum contents of 190 ppm P and 0.6% reactive Fe (Pit et al., 2017). Large-scale pyrite oxidation and geological differences in the parent material of the mega beach nourishment are responsible for the high contents of P and Fe oxides. These geological differences, which include Holocene and Pleistocene sediments, were visible in the sand pit that supplied the material to create the Sand Engine: near the seafloor there were contents of over 300 ppm P and 1% reactive Fe that declined rapidly with depth, to only 50 ppm P at the bottom of the sand pit. Additionally, the fine fraction (<150 µm) contained more P and Fe compared to the coarse (150–2000 µm) fraction.

When pH > 6 and the carbonate content is high, it is assumed that P is mainly bound to Ca as Ca-phosphates (Kooijman et al., 1998). However, Pit et al. (2017) showed a high correlation between Fe and P at the Sand Engine, where the expected P speciation may be changed to Fe-bound P. Given that sand nourishments are crucial for the coastal management of the Netherlands, a potentially high content of P on Dutch beaches can be expected (Pit et al., 2017; Stuyfzand et al., 2012). This may create a large source of additional P that may enter the dune ecosystems. To date, it is unclear whether the P associated with the sand nourishment material enters the dune system and alters the P availability of the dune soil, thereby possibly having consequences for the dune vegetation.

We set out to study the sand and vegetation characteristics of the beach and dune area at two different locations: (1) the Sand Engine, a mega sand nourishment without a dynamic dune system and (2) a traditional sand nourishment with a dynamic dune system through the creation of five notches in the foredunes perpendicular to the coastline. From a nature conservation perspective, a dynamic dune system is preferred over a fixed dune system to facilitate natural transport of beach sand into the dunes, thereby providing the dune area with new calcareous material, limiting acidification and stimulating dune development (Arens, 2001; Arens et al., 2013b).

With the Netherlands being dependent on sand nourishments for their coastal management, a potentially high concentration of P on the beach can be expected (Pit et al., 2017; Stuyfzand et al., 2012), creating large source of additional P that may enter the dune ecosystems. Therefore, the aim of this study is to investigate if the P content of sand nourishment material enters the dune system and whether this material alters the P availability of the soil and as a result affect the N:P ratio of dune vegetation.

We hypothesized that in the presence of a dynamic dune system and nourishment material on the beach, P would be transported to the dune area via eolian processes. As the foredunes are the first dunes to receive nourishment sand, the influence of P is assumed to be highest here. We also hypothesized that on the foredunes of the mega nourishment, the P content is higher than on the foredunes of the traditional nourishment, because of the larger volume of sand and higher P content in the fine fraction that can be transported by wind and create local enrichment with P. However, at the traditional nourishment site, the dynamic dune system is expected to transport more fresh material from the beach to the inner dunes than at the fixed dune system at the mega nourishment, where no dynamic dune system is present.

At the mega sand nourishment, the P of beach material and transported sand is expected to be more available for plants compared to the traditional nourishment site. The expected predominance of Fe-

bound P at the mega nourishment may cause a decrease in sensitivity to pH changes, as Ca-bound P dissolves more easily due to decalcification (Golterman, 1988; Kooijman et al., 1998). However, the total P content is expected to be higher at the mega nourishment, which will create an overall higher P availability. Since N deposition along the Dutch coast shows no marked gradients (Kooijman et al., 2012; Stolk et al., 2009), we expected the N:P ratios would be lower in plants growing in the foredunes than in plants growing further inland on the dunes at the mega nourishment. For the traditional nourishment site we expected a more uniform N:P ratio in the plants in the dune area because of a dynamic dune system behind the traditional nourishment.

## 2. Material & methods

### 2.1. Field sites

We compared two sites (52°02'40.3"N; 4°10'59.4"E and 52°25'34.3"N; 4°33'25.2"E) along the Dutch coast (Fig. 1), located in a carbonate-rich environment (>2% CaCO<sub>3</sub>; Eisma, 1968). Each site has a dune area with foredunes (developed dunes formed with pioneer plant communities), white dunes (where seawater effects are limited and no soil has yet developed), and grey dunes (formed behind the beach ridge where wind dynamics are low and a soil profile has formed; Hesp, 2002; Janssen and Schaminée, 2003; LNV, 2004).

The first site is the Sand Engine pilot project (SE) created in 2011, a mega sand nourishment of 21.5 million m<sup>3</sup> of sand that is developing and expected to result in a wider beach along a 10–20 km stretch of the Dutch coastline over a 20-years period, as well as a beach area gain of 200 ha (Stive et al., 2013). The Sand Engine strategy is offensive: not much sand is transferred toward the dune area because that is shielded by the foredunes (Vertegaal et al., 2016), which are referred to as fixed foredunes. As a result, grey dunes will not be replenished and long-term effects are unclear. The second site is an irregular replenished area by means of foreshore nourishments and beach nourishments in the Zuid-Kennemerland National Park near Haarlem, the Netherlands. Unlike the Sand Engine, a sand sharing system was created in 2012 and 2013, the so-called project Northwest Nature Core (NWC) (Kuipers, 2014; Kuipers et al., 2016). A total of 140,000 m<sup>3</sup> of sand was removed from the foredunes in five parallel notches perpendicular to the coastline, initiating active erosion zones on either side of the notches (Ruessink et al., 2018), which are referred to as dynamic

foredunes. As a result, a long transport gradient was created, where sand is expected to replenish the white dunes and part of the grey dunes, in order to counteract acidification of dune sands (Kuipers et al., 2016).

### 2.2. Sampling

Fieldwork was carried out in summer 2016. At each of the two sites (SE and NWC), the incoming sand, dune and beach sand, and vegetation were sampled. The sampling points (Fig. 1) were along two transects in NWC and four transects in SE. The transects were selected so as to include three existing sand traps per transect (installed to monitor eolian sand transport in the white and grey dunes based on Leatherman (1978)); at NWC there were two additional sand traps located in dune slacks (Arens et al., 2016; Vertegaal et al., 2016). These sand traps have been sampled every other week since 2012 at SE and since 2013 at NWC, for analysis. The samples from the sand traps contained 0–702 g of sand. To determine the characteristics of the transported sand, sand from sand traps in the dune area was collected.

The study sites were divided into four different zones: beach (zone 1), frontal foredunes (zone 2), foredunes crest (zone 3), and inner dunes (zone 4) (Figs. 1 and 2). Sand traps were present only in zones 3 and 4. At NWC, the dynamic site with traditional beach nourishment, the sand traps yielded a total of 33 samples, with 2 to 6 sand trap samples per sampling date, and sampling dates ranging from October 8th, 2013 till March 22nd, 2016. However, the sand traps contained very little sand in the non-dynamic mega nourishment site SE because of the limited transport of sand from the beach to the dune area. Therefore, although the superficial sand of the beach and foredunes was sampled in both sites, we report the sand trap results for NWC only. Unfortunately, the trapped sand samples did not contain enough material for all measurements. Sometimes only one sample was available; in such cases, no standard deviation could be reported. There were eight trapped sand samples available for NWC, six of which were taken on the same date (Dec 01 2015). The remaining three samples are from the same sand trap in zone 3 but were sampled on different dates (Dec 01 2015, Aug 05 2014 and Dec 09 2014).

At each sampling point in SE and NWC, sand and vegetation were sampled. For the superficial sand samples, a geological sample bag was filled with approximately 400 g of sand scooped from approximately the top 5 cm with a trowel. Additionally, a stainless steel ring

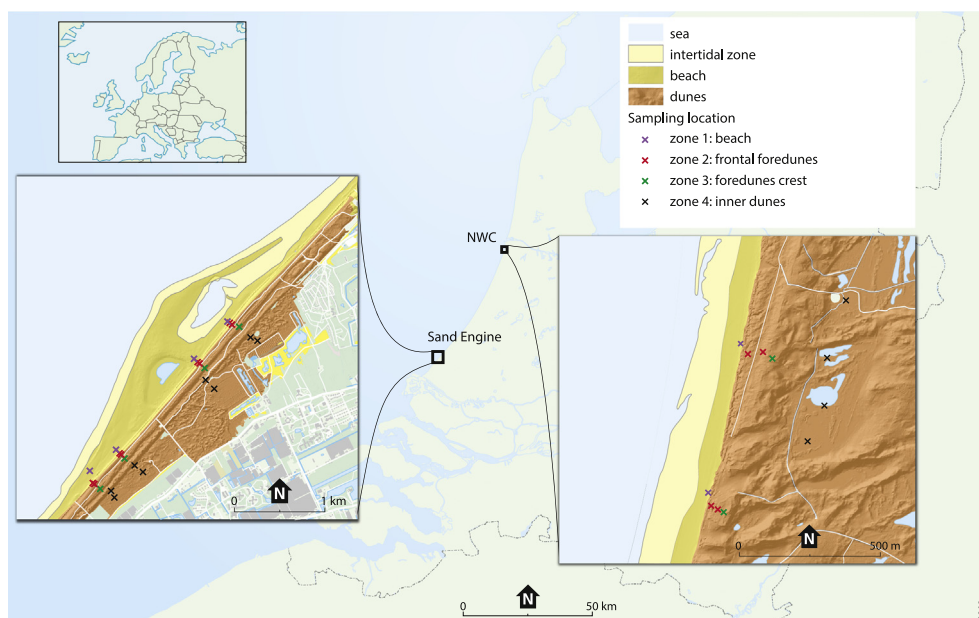
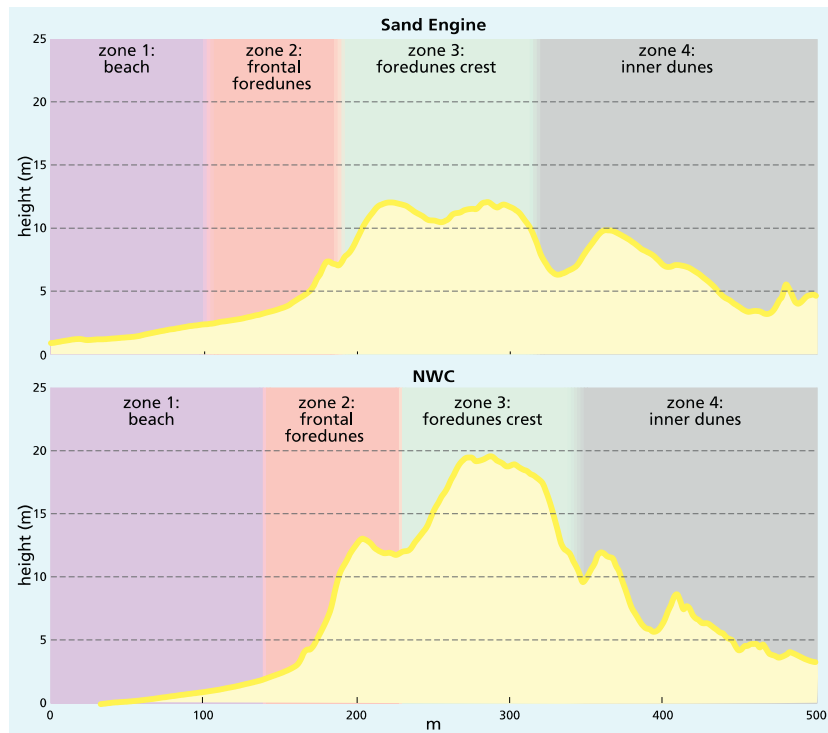


Fig. 1. The locations of the sampling sites. The locally concentrated sand nourishment the Sand Engine (SE) and the dynamic dune area project, Northwest Nature Core (NWC).



**Fig. 2.** For a clear overview of the different zones, the average height compared to sea level of the two study locations Sand Engine and NWC. Data was retrieved from the Ministry of Infrastructure and Water Management (2014 for the Sand Engine and 2018 for NWC). Measurements start at the waterline with 0 m being the sea level and end 500 m inland covering the four zones where samples were taken.

with a diameter of 5.3 cm and a length of 5 cm was filled and closed with plastic caps for the estimation of the bulk density. To obtain an indication of the plant-available P at each site, two plant species were sampled: *Ammophila arenaria* (Marram grass) and *Carex arenaria* (Sand sedge) and their N and P contents measured. Both plant species are typical for temperate coastal regions. *A. arenaria* is a sand stabilizer that occurs on growing coastal sand dunes (Barr and McKenzie, 1976; Doing, 1985; Van der Laan et al., 1997), where it forms new roots in newly deposited windblown beach sand (Van der Stoel et al., 2002). *C. arenaria* grows in patches on fixed grey dunes (Noble et al., 1979). Due to the different location of these species, it is possible to estimate the impact of nourishment material in different parts of the dune area. Depending on the sampling point, both or only one of the two plant species were found. At SE, *A. arenaria* was found in each of the four zones, whereas *C. arenaria* was only present in zones 3 and 4. At NWC, *A. arenaria* was present in zones, 1, 2, and 3, and *C. arenaria* was present in zones 2, 3 and 4, but only one sample of *C. arenaria* was found in zone 2.

At each location, approximately 30 g of living aboveground biomass consisting predominantly of leaves was collected per plant species present. The sample was put in a paper bag which was placed open on a heater the same day for pre-drying. A week after completing the fieldwork, vegetation samples were dried in an oven at 70 °C for 48 h before analysis.

### 2.3. Chemical and grain size analyses

Chemical analyses were performed on both vegetation and sand samples. All sand samples were dried in the oven at 105 °C for at least 24 h. Using a subsample, the grain size distribution was measured with a laser diffraction analyzer (Helos KR Sympatic) after pre-treatment in accordance to Konert and Vandenberghe (1997) to reduce aggregation of the soil particles and remove organic material. The pH of the sand was measured in accordance with NEN 5750 (1989). For the estimation of the bulk density, the dry weight of the sand samples in the stainless steel ring was measured and divided by the volume of

the stainless steel ring. Since solid mass density varied at both sites, the chemical contents of the sand were recalculated in this paper to g/m<sup>2</sup> per 5 cm depth of soil to enable true comparison of the characteristics.

Additionally, 12 sand samples per study site (sampling points on the outermost transects of SE and all sampling points at NWC) were sieved to obtain two fractions: <250 μm (>60 mesh) and 250–2000 μm (10–60 mesh). The fine fraction of <250 μm is more likely to reach the inner dunes than the large fraction of 250–2000 μm (Abuodha, 2003; Van Buuren, 2017). The solid P speciation was characterized for the two fractions using a sequential extraction method (Ruttenberg, 1992). The extraction procedure involved five steps to obtain (1) exchangeable or loosely sorbed P, (2) easily reducible or reactive ferric Fe–P (Fe-bound P), (3) amorphous apatite and carbonate P (Ca-bound P), (4) crystalline apatite and detrital P (detrital P), and (5) organic P (Saaltink et al., 2016). In addition to characterizing solid P speciation, the content of Fe oxides was estimated by using the ICP-OES to analyze the extractants for the Fe-bound P step on Fe as well. Additionally, total P was calculated by totaling the sequentially extracted amounts of P.

Next to a sequential extraction, the plant-available P content in the sand was estimated using a cation and anion exchange membrane strip (CAEM). The method used is based on Saggari et al. (1990), but modified according to Myers et al. (2005) and Kroth et al. (2016). First, we used an indication of total P from the sequential extraction to determine the maximum size of each membrane strip, which was done via the sequential extraction. The CEM and AEM sheets were cut into strips of 8.3 × 2.1 cm so that the surface would be large enough to retain the P that became available from the sand and the strips would fit easily into a 50 mL Greiner tube. Both strips were placed in a Greiner tube which was then filled with 0.5 M NaHCO<sub>3</sub> solution to saturate the strips for 24 h. The strips were then rinsed with ultrapure water and replaced in the Greiner tube together with approximately 1 g of sand and 10 mL of ultrapure water, before being put on a reciprocating shaker at 265 rpm. After 24 h, the strips were removed from the

Greiners with plastic tweezers and any grains were washed off carefully using a siphon with ultrapure water. The strips were then placed in Greiners with 10 mL of 0.5 HCl solution and placed on the reciprocating shaker for 90 min at 265 rpm. After the strips had been removed, the solution was measured on the ICP-OES to estimate the content of plant-available P. A test with 4 different dilutions of a NaPO<sub>4</sub> stock solution using the same procedure confirmed the accuracy of this method: average recovery was 98%.

For the other sand analyses, both fractions were ground to <2 µm with a Herzog HP-PA grinding machine or by hand when not enough material was available. The fractions were analyzed for total C and N contents with a CN elemental analyzer (Fisons NA 1500 NCS). The aqua regia digestion method (Houba et al., 1995), was used to obtain the pseudo-total elemental contents (Chen and Ma, 2001), which were analyzed with inductively coupled plasma optical emission spectrometry (ICP-OES) and mass spectrometry (ICP-MS). P contents estimated with the aqua regia digestion method were used to check the accuracy of the total P values calculated from the sequential extraction. On average, the total P values from the sequential extraction were 92% of the P contents derived from the aqua regia digestion method ( $R^2 = 0.55$ ).

The vegetation samples were ground with a Foss Cyclotec 1093 and a subsample was destroyed in a microwave (Milestone MLS 1200 Mega) in a Teflon® destruction vial containing 5 mL concentrated nitric acid (65% HNO<sub>3</sub>) and 2 mL hydrogen peroxide (30% H<sub>2</sub>O<sub>2</sub>). After destruction, analyses were performed with an inductively coupled plasma emission spectrometer (ICP-OES ICAP 6300, Thermo Fisher Scientific of, ARCOS MV, Spectro) for the elements P, K, Ca, Mg, Mn, Zn, Si, and Al. Additionally, the ground samples were used to measure total C and total N on a CNS element analyzer (Carlo Erba-Thermo Fisher Scientific EA NA 1500 and EA100).

#### 2.4. Statistical analyses

To test whether there were significant differences between the mega nourishment site without a dynamic dune system and the traditional nourishment site with a dynamic dune system, statistical analyses were carried out for both the sand dataset – including superficial sand and sand trap data – and the plant dataset. SPSS was used to perform the statistical analyses. Data were tested for normality and homogeneity of variance, using the Shapiro-Wilk and Levene tests (Field, 2009).

For the plant tissue concentrations, the distribution for *C. arenaria* and *A. arenaria* per location was normal and variances did not differ significantly. Therefore, an independent *t*-test was chosen to determine if there is a significant difference between the zones and/or two sites. The same test was performed for the comparison of the two sand fractions, which showed a normal distribution and no significant difference in the homogeneity of variances per site. For the comparison of the four zones, the data was normal distributed, but homogeneity of variance differed and, therefore, an independent ANOVA with a Games Howell post hoc test was chosen, because this test works best when sample size per groups are unequal and population variances differ (Field, 2009). For the superficial sand dataset, a three-way independent ANOVA (Kutner et al., 2010) with general linear models was used to

determine the main and interaction effects of the three independent variables: site (SE and NWC), zone (1 to 4), and sand fraction (<250 µm and 250–2000 µm). Differences were considered to be statistically significant at  $p \leq 0.05$ .

Additionally, Canoco 5 was used for multidimensional scaling and to estimate links between sand properties and vegetation characteristics (Ter Braak and Šmilauer, 2012). An unconstrained principal component analysis (PCA) based on the correlation matrix was performed on the sand dataset. In order to be able to compare the findings of the superficial sand samples with the trapped sand samples in a PCA, the values were adjusted to  $\text{g m}^{-2} (5 \text{ cm})^{-1}$ , using the average bulk density of superficial sand samples from zones 1 and 2 for the trapped sand samples.

### 3. Results

#### 3.1. Superficial sand at the beach and dune area

##### 3.1.1. General characteristics

The general characteristics of superficial sand at the two study sites are shown in Table 1 for each zone: (1) beach, (2) frontal foredunes, (3) foredunes crest, and (4) inner dunes. At SE, the mega nourishment site with fixed foredunes, the grain size distribution was significantly coarser in zones 1 to 3 than in zone 4, as can be seen from the mean values of the median grain size D50 and the fraction < 250 µm. The pH was also significantly higher for zones 1–3 than for zone 4, with values of 8.0 or higher versus 6.0.

At NWC, the traditional nourishment site with dynamic foredunes, the variables D50 and <250 µm fraction indicating no significant differences in grain size among the zones. However, pH was significantly higher for zone 3 than for zone 4, with values above 8 versus 7.5. The bulk density (Table 1) was significantly higher for zones 1 to 3 compared to zone 4.

##### 3.1.2. Three-way independent ANOVA on superficial sand

For the superficial sand, the interaction effect between the chemical characteristics and the three independent variables, site (SE and NWC), zone (1–4) and sand fraction (<250 µm and 250–2000 µm), was examined with a three-way independent ANOVA (Table 2) using the adjusted sand characteristics ( $\text{g m}^{-2} (5 \text{ cm})^{-1}$ ). Significance holds when a response variable is dependent on 1, 2 or 3 independent variables. The results showed that only CAEM-available P and aqua regia content of K were affected when the site, zone and sand fraction were considered. There was no three-way interaction for the other response variables. For the majority of the response variables, significant differences were observed when the zone and fraction were involved, except in the case of the response variables Fe-bound P, organic P, Fe oxides, and available P. This suggests that at each site, there were significant differences between the zones and the sand fractions in the binding of P related to Ca, other inorganic minerals, and exchangeable P. Here, this may not result in P becoming available at significantly higher or lower contents. To conclude, there were no significant differences between NWC and SE, except for the CAEM-available P fraction and aqua regia content of K. Most of the response variables were dependent on two independent variables: the zone and the sand fraction.

**Table 1**

General characteristics of the sand sampled from the Sand Engine (SE) and North West Core (NWC) per zone, with means and standard deviations (st. dev). Groups found to differ significantly (Games Howell post doc,  $p < 0.05$ ) from other groups are indicated with different letters.

|                                  | SE           |               |              |             | NWC          |              |               |              |
|----------------------------------|--------------|---------------|--------------|-------------|--------------|--------------|---------------|--------------|
|                                  | Zone 1       | Zone 2        | Zone 3       | Zone 4      | Zone 1       | Zone 2       | Zone 3        | Zone 4       |
| n                                | 4            | 8             | 4            | 8           | 2            | 4            | 2             | 4            |
| D50 (µm)                         | 376 (25) b   | 327 (31) b    | 317 (35) b   | 231 (27) a  | 275 (23) a   | 264 (25) a   | 299 (47) a    | 215 (12) a   |
| <250 µm (%)                      | 17 (5) a     | 25 (7) a      | 26 (11) a    | 61 (11) b   | 41 (9) a     | 45 (10) a    | 33 (16) a     | 68 (7) a     |
| pH                               | 8.6 (0.1) c  | 8.3 (0.3) bc  | 8.0 (0.2) b  | 6.0 (0.6) a | 8.6 (0.2) ab | 8.3 (0.4) ab | 8.2 (0.03) bc | 7.5 (0.2) a  |
| Bulk density ( $\text{g/cm}^3$ ) | 1.6 (0.07) c | 1.5 (0.06) bc | 1.4 (0.06) b | 1.1 (0.2) a | 1.5 (0.07) b | 1.5 (0.07) b | 1.4 (0.2) bc  | 0.9 (0.1) ac |

**Table 2**  
Results of the three-way independent ANOVA for the sand characteristics with the independent variables site, zone, and sand fraction. The results are either significant when <0.05 or not significant (n.s.).

|                         |                | Site  | Zone  | Sand fraction | Site · zone | Site · sand fraction | Zone · sand fraction | Site · zone · sand fraction |
|-------------------------|----------------|-------|-------|---------------|-------------|----------------------|----------------------|-----------------------------|
| CN analyzer             | N              | n.s.  | 0.000 | n.s.          | n.s.        | n.s.                 | 0.001                | n.s.                        |
|                         | C              | n.s.  | 0.002 | n.s.          | n.s.        | n.s.                 | 0.000                | n.s.                        |
| Sequential P extraction | Exchangeable P | n.s.  | 0.000 | 0.012         | 0.013       | n.s.                 | 0.000                | n.s.                        |
|                         | Fe-bound P     | n.s.  | 0.005 | 0.001         | 0.036       | 0.037                | n.s.                 | n.s.                        |
|                         | Ca-bound P     | n.s.  | 0.025 | 0.000         | n.s.        | n.s.                 | 0.000                | n.s.                        |
|                         | Detrital P     | n.s.  | 0.001 | 0.000         | n.s.        | n.s.                 | 0.007                | n.s.                        |
|                         | Organic P      | n.s.  | 0.002 | n.s.          | n.s.        | n.s.                 | n.s.                 | n.s.                        |
|                         | Total P        | n.s.  | n.s.  | 0.000         | n.s.        | 0.011                | 0.000                | n.s.                        |
|                         | Fe oxides      | n.s.  | n.s.  | 0.032         | n.s.        | n.s.                 | n.s.                 | n.s.                        |
| CAEM                    | Available P    | 0.029 | 0.000 | n.s.          | 0.000       | n.s.                 | n.s.                 | 0.02                        |
| Aqua regia              | Ca             | n.s.  | 0.016 | 0.000         | 0.022       | 0.01                 | 0.000                | n.s.                        |
|                         | Fe             | n.s.  | n.s.  | 0.006         | 0.016       | 0.000                | 0.001                | n.s.                        |
|                         | K              | 0.002 | 0.001 | 0.000         | 0.007       | 0.000                | 0.000                | 0.012                       |

### 3.1.3. Specific sand characteristics

Specific sand characteristics are shown in Table 3 and the P bindings with the highest P contents are presented in Fig. 3 for coarse and fine sand fractions (250–2000 µm and <250 µm) per site and for each zone. First, the variation of the P content is described per zone, followed by a comparison of the P bindings of the two sand fractions at SE and NWC.

The chemical characteristics showed that the P content did not vary much for the different zones and fractions at SE and NWC, except for lower total P values for the fine fraction in zone 4 at NWC, which was significantly different from zone 1–3. Interestingly, the coarse fraction at NWC showed a higher total P content in zone 4 than in the other zones and much higher compared to SE, but the difference was not significant because the standard deviation was large. Additionally, the

coarse fraction showed a very high content of C in zone 4 in both SE and NWC. Building up of organic matter in zone 4 may account for the (slightly) higher results for the coarse fraction, especially at NWC where sampling plots were in dune slacks.

Comparison of the two sand fractions revealed a similar variation in the P bindings in zone 1–3 for both SE and NWC. For the fine fraction at SE, most (41–62%) of the total P was present in so-called detrital P, which was significantly higher compared to the coarse fraction. In the coarse fraction most (38–58%) of the total P was Fe-bound P, which was significantly higher compared to the fine fraction, except for zone 2, where detrital P was slightly higher than Fe-bound P. Additionally, of the total P, Ca-bound P occurred significantly more in the fine fraction than in the coarse fraction (0.1–21% versus 0.02–0.06%). Therefore,

**Table 3**  
Chemical characteristics of the sand samples at Sand Engine (SE) and North West Core (NWC) per zone, with the means and standard deviations (st.dev). Groups found to differ significantly (Games Howell post doc,  $p < 0.05$ ) from other groups are indicated with different letters.

| SE                      | n              | 250–2000 µm                           |              |              |               | < 250 µm                              |               |                |              |
|-------------------------|----------------|---------------------------------------|--------------|--------------|---------------|---------------------------------------|---------------|----------------|--------------|
|                         |                | Zone 1                                | Zone 2       | Zone 3       | Zone 4        | Zone 1                                | Zone 2        | Zone 3         | Zone 4       |
|                         |                | 2                                     | 4            | 2            | 4             | 2                                     | 4             | 2              | 4            |
|                         |                | $\text{g m}^{-2} (5 \text{ cm})^{-1}$ |              |              |               | $\text{g m}^{-2} (5 \text{ cm})^{-1}$ |               |                |              |
| CN analyzer             | N              | 1.6 (0.1) a                           | 2.4 (1.9) a  | 11 (0.3) b   | 138 (124) ab  | 4.4 (0.8) a                           | 4.3 (5.1) a   | 20 (8.5) a     | 32 (43) a    |
|                         | C              | 120 (74) a                            | 127 (67) a   | 238 (20) a   | 2014 (1859) a | 1097 (431) a                          | 631 (277) a   | 836 (212) a    | 482 (427) a  |
| Sequential P extraction | Exchangeable P | 0 (0) a                               | 0 (0) a      | 0 (0) a      | 0.2 (0.1) a   | 0 (0) a                               | 0 (0) a       | 0 (0) a        | 0.1 (0.12) a |
|                         | Fe-bound P     | 3.1 (0.5) a                           | 1.5 (0.6) a  | 3.1 (0.9) a  | 4.7 (3.1) a   | 2.4 (0.3) a                           | 1.5 (0.4) a   | 2.4 (0.3) a    | 3.9 (1.2) a  |
|                         | Ca-bound P     | 0.4 (0.3) a                           | 0.1 (0.1) a  | 0.2 (0.1) a  | 0.3 (0.2) a   | 2.1 (0.2) a                           | 2.1 (0.2) a   | 2.3 (1.3) a    | 0.9 (0.8) a  |
|                         | Detrital P     | 2.7 (1) a                             | 1.7 (0.2) a  | 1.9 (0.1) a  | 1.7 (0.7) a   | 8.1 (2.5) a                           | 5.7 (2.2) a   | 8.1 (3.7) a    | 4.1 (1.6) a  |
|                         | Organic P      | 0.5 (0.2) a                           | 0.6 (0.5) a  | 0.5 (0.2) a  | 1.2 (1.1) a   | 0.4 (0.02) a                          | 0.5 (0.8) a   | 0.7 (0.3) a    | 0.9 (0.5) a  |
|                         | Total P        | 6.6 (1.9) a                           | 3.9 (0.5) a  | 5.6 (1) a    | 8.1 (3.1) a   | 13 (2.4) a                            | 9.8 (3.5) a   | 14 (5) a       | 9.8 (2.2) a  |
|                         | Fe oxides      | 119 (16) a                            | 64 (16) a    | 106 (38) a   | 62 (41) a     | 107 (0.8) b                           | 75 (15) ab    | 82 (15) ab     | 49 (6.4) a   |
| CAEM                    | Available P    | 0.2 (0) a                             | 0.1 (0.02) a | 0.3 (0.04) a | 1.5 (0.7) a   | 0.2 (0.1) a                           | 0.2 (0.04) a  | 0.4 (0.1) ab   | 0.8 (0.2) b  |
| Aqua regia              | Ca             | 206 (42) a                            | 211 (60) a   | 296 (96) a   | 115 (72) a    | 3939 (1524) b                         | 2338 (878) ab | 2367 (1037) ab | 250 (323) a  |
|                         | Fe             | 210 (30) a                            | 182 (18) a   | 234 (60) a   | 182 (63) a    | 532 (97) a                            | 345 (68) a    | 378 (120) a    | 215 (20) a   |
|                         | K              | 27 (2.7) a                            | 30.6 (2.4) a | 34 (6.4) a   | 33 (6) a      | 93 (16) a                             | 63 (14) a     | 61 (10) a      | 39 (4.1) a   |

| NWC                     | n              | 250–2000 µm                           |              |              |               | < 250 µm                              |              |              |             |
|-------------------------|----------------|---------------------------------------|--------------|--------------|---------------|---------------------------------------|--------------|--------------|-------------|
|                         |                | Zone 1                                | Zone 2       | Zone 3       | Zone 4        | Zone 1                                | Zone 2       | Zone 3       | Zone 4      |
|                         |                | 2                                     | 4            | 2            | 4             | 2                                     | 4            | 2            | 4           |
|                         |                | $\text{g m}^{-2} (5 \text{ cm})^{-1}$ |              |              |               | $\text{g m}^{-2} (5 \text{ cm})^{-1}$ |              |              |             |
| CN analyzer             | N              | 2.2 (0.1) a                           | 5.1 (6.3) a  | 7.8 (4.4) a  | 217 (108) a   | 19 (12) a                             | 38 (32) a    | 82 (89) a    | 43 (34) a   |
|                         | C              | 204 (41) a                            | 248 (100) a  | 260 (58) a   | 3314 (1709) a | 701 (249) a                           | 833 (508) a  | 1361 (998) a | 897 (249) a |
| Sequential P extraction | Exchangeable P | 0 (0) a                               | 0 (0) a      | 0 (0) a      | 0.5 (0.1) b   | 0 (0) a                               | 0 (0) a      | 0 (0) a      | 0.1 (0.1) a |
|                         | Fe-bound P     | 3.7 (0.04) a                          | 4 (0.3) a    | 4.2 (0.8) a  | 11 (11) a     | 2.1 (0.2) ab                          | 2.4 (0.1) ab | 3.3 (0.9) b  | 1.6 (0.3) a |
|                         | Ca-bound P     | 0.1 (0) a                             | 0.3 (0.2) ab | 0.2 (0) b    | 6.3 (11) ab   | 1.6 (0.2) a                           | 2.2 (0.7) a  | 2.1 (0.6) a  | 0.8 (0.3) a |
|                         | Detrital P     | 1.9 (0.4) a                           | 2.2 (0.6) a  | 1.3 (0.2) a  | 1.5 (0.2) a   | 6.3 (0.7) b                           | 5.3 (1.6) ab | 6.6 (1.2) ab | 2.3 (0.8) a |
|                         | Organic P      | 0.5 (0.1) a                           | 0.8 (0.2) a  | 0.5 (0.04) a | 3.4 (2) a     | 0.4 (0) a                             | 0.3 (0.3) a  | 0.3 (0.4) a  | 0.8 (0.3) a |
|                         | Total P        | 6.3 (0.3) a                           | 7.3 (0.6) a  | 6.2 (0.5) a  | 23 (23) a     | 10.4 (0.8) b                          | 10 (1.8) b   | 12 (0.6) b   | 5.5 (0.7) a |
|                         | Fe oxides      | 115 (0.9) a                           | 121 (11) a   | 117 (10) a   | 208 (193) a   | 76 (5.8) a                            | 81 (7) a     | 98 (8) a     | 71 (52) a   |
| CAEM                    | Available P    | 0.2 (0.04) a                          | 0.2 (0.1) a  | 0.3 (0.1) a  | 0.8 (1.2) a   | 0.2 (0.01) a                          | 0.2 (0.1) a  | 0.4 (0.2) a  | 0.5 (0.2) a |
| Aqua regia              | Ca             | 574 (157) a                           | 496 (128) a  | 425 (45) a   | 1448 (1599) a | 2108 (110) a                          | 1860 (78) a  | 1992 (113) a | 766 (634) a |
|                         | Fe             | 207 (13) a                            | 231 (19) a   | 250 (52) a   | 335 (223) a   | 254 (12) a                            | 256 (8) a    | 288 (28) a   | 172 (52) a  |
|                         | K              | 14 (0.7) a                            | 16 (0.7) a   | 15 (3.3) a   | 21 (4.7) a    | 33 (1) b                              | 31 (1) b     | 33 (5.5) ab  | 21 (3.5) a  |

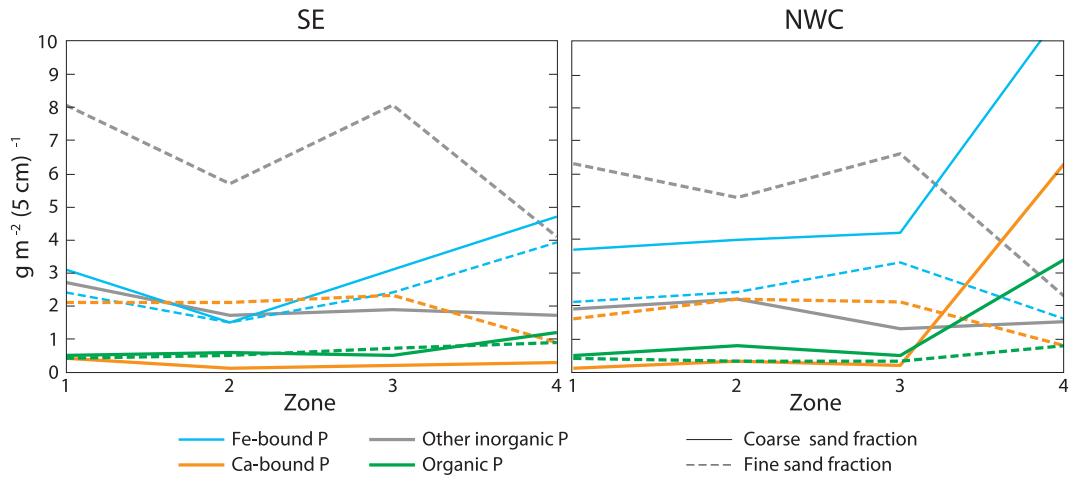


Fig. 3. The P contents of the P bindings Fe-bound P, Ca-bound P, other inorganic P and organic P for coarse and fine sand fractions per site and for each zone.

three main P bindings can be distinguished for the fine fraction (detrital P, Fe-bound P, and Ca-bound P) and two main P bindings for the coarse fraction (Fe-bound P and detrital P). Additionally, total P showed to be significantly higher for the fine fraction compared to the coarse fraction.

The difference between the two sand fractions was not mirrored by the CAEM-available P, the content of which was similar for both fractions, although the fine fraction had a significantly higher content in zone 4 than in zones 1 and 2. Aqua regia contents of Ca showed a significant difference for the fine fraction, with much lower values for zone 4 than for zone 1. The pattern for zones 1–3 at NWC was similar to that found at SE. The total P of the fine fraction mainly comprises detrital P (53–61%), followed by Fe-bound P (20–28%), and then Ca-bound P (15–22%). These variables showed a significant difference between the fine and the coarse fraction. The total P of the coarse fraction contained most of the Fe-bound P (55–58%); detrital P accounts for 21–30%. As a result, the P differences were more pronounced between the zones (zone 1–3 versus zone 4) and between the fractions rather than between the sites, which confirmed the results of the three-way independent ANOVA.

3.2. Sand traps

The chemical characteristics of the samples from the trapped sand (present in the dune area of NWC, zones 3 and 4) are shown in Table 4 and the associated grain size distribution is presented in Table S1. Finer sand was transported to zone 4, where the median grain size distribution was slightly lower for the trapped sand in zone 4 (202 μm) than that for

the trapped sand in zone 3 (213 μm). Also, the fraction <250 μm was higher for zone 4 than for zone 3 (80% versus 71%). Therefore, most of the trapped sand toward zone 3 and 4 was fine sand with <250 μm.

The chemical characteristics of the fine fraction showed that the total P content of the trapped sand in zone 3 and 4 (Table 4) was slightly higher than that of the fine fraction of the superficial sand samples of zones 1 to 4 (Table 3). The main P bindings per zone were similar for both types of samples: most of the total P comprises detrital P (55–56%), then Ca-bound P (25%) and Fe-bound P (17%). It seems that compared to the other P bindings, higher contents of so-called detrital P and Ca-bound P were being transported toward the dune area of NWC. To further investigate the relations between the two types of samples, a PCA was performed (see Section 3.3). The total P content of the coarse fraction of the sand trap samples (Table 4) was also slightly higher than that of the superficial sand samples (Table 3). Most of the total P in coarse sand was Fe-bound P (56–64%), with detrital P accounting for a further 27–30%; a similar pattern was found for the coarse fraction of the superficial sand samples. CAEM-available P contents were similar for both fractions of the trapped sand. The values were slightly lower or comparable to the superficial sand samples of zone 1 and 2, and lower than the superficial samples of zone 3 and 4.

Other significant differences for the fine fraction of sand trap samples were found for the content of Fe oxides: it was higher in zone 4 than in zone 3. For the coarse fraction, the aqua regia contents of Ca, Fe, and K were significantly higher in zone 4 than in zone 3, with Ca being especially high, perhaps because of the presence of shell material.

Table 4

Chemical characteristics and their means and standard deviations (st.dev) of the samples collected from the sand traps at North West Core (NWC). Significant differences (independent t-test, p < 0.05) between the two zones are indicated by \*. When there was only one sample in a zone, the lack of a standard deviation is indicated by (-).

| NWC                     |                | 250–2000 μm                            |               | Sig. | <250 μm       |  | Sig. |
|-------------------------|----------------|--|---------------|------|---------------|--|------|
|                         |                | Zone 3                                 | Zone 4        |      | Zone 3        | Zone 4                                 |      |
|                         |                | Mean (st.dev)                          | Mean (st.dev) |      | Mean (st.dev) | Mean (st.dev)                          |      |
|                         |                | g m <sup>-2</sup> (5 cm) <sup>-1</sup> |               |      |               | g m <sup>-2</sup> (5 cm) <sup>-1</sup> |      |
| CN analyzer             | N              | 1152 (94)                              | -             |      | 18 (15)       | 11 (1.1)                               |      |
|                         | C              | 283 (236)                              | -             |      | 643 (93)      | 683 (34)                               |      |
| Sequential P extraction | Exchangeable P | 0.3 (0.5)                              | 0.0 (0.0)     |      | 0.1 (0.1)     | 0.0 (0.0)                              |      |
|                         | Fe-bound P     | 4.9 (0.5)                              | 5.4 (1.1)     |      | 2.3 (0.1)     | 2.3 (0.2)                              |      |
|                         | Ca-bound P     | 0.5 (0.4)                              | 0.3 (0.1)     |      | 3.2 (1.0)     | 3.3 (0.7)                              |      |
|                         | Detrital P     | 2.6 (1.8)                              | 2.3 (0.3)     |      | 7.3 (1.4)     | 7.2 (1.8)                              |      |
|                         | Organic P      | 0.5 (0.4)                              | 0.4 (0.6)     |      | 0.3 (0.3)     | 0.4 (0.1)                              |      |
|                         | Total P        | 8.8 (2.5)                              | 8.4 (1.6)     |      | 13 (1.8)      | 13 (1.2)                               |      |
| CAEM                    | Fe oxides      | 141 (16)                               | 169 (39)      |      | 78 (7.5)      | 92 (3.2)                               | *    |
|                         | Available P    | 0.1 (0.0)                              | -             |      | 0.2 (0.2)     | 0.1 (0.0)                              |      |
| Aqua regia              | Ca             | 974 (619)                              | 5465 (-)      | *    | 1968 (251)    | 2158 (80)                              |      |
|                         | Fe             | 263 (30)                               | 461 (-)       | *    | 274 (15)      | 292 (5.6)                              |      |
|                         | K              | 36 (9.5)                               | 72 (-)        | *    | 52 (4.9)      | 55 (2.6)                               |      |



The N content in zone 3 was also exceptionally high. As a result, for both fractions the total P content was slightly higher for trapped sand compared to the superficial samples, with comparable main P bindings and a similar or slightly lower available P content. Because the majority of the trapped sand at NWC is <250  $\mu\text{m}$ , higher contents of inorganic P and Ca-bound P were being transported toward the dune area.

### 3.3. Relations between zones and fractions, and trapped sand versus superficial sand

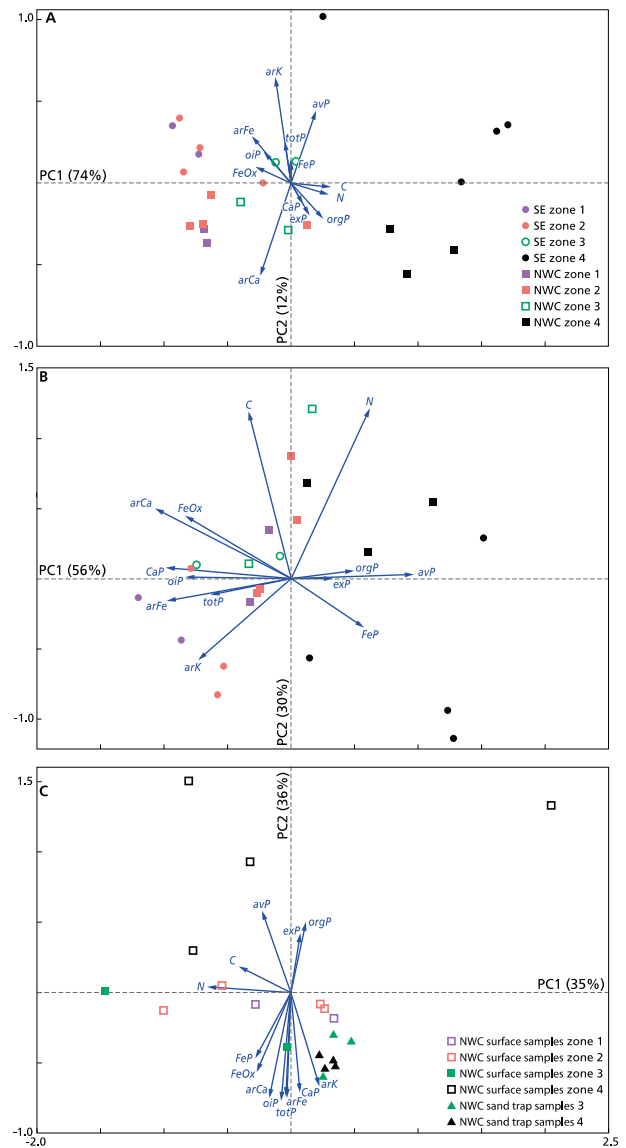
A PCA was performed (Fig. 4) to further investigate the connections between zones for each fraction as well as associations between the geochemical composition of fine superficial sand and sand trap samples. Fig. 4 shows ordination diagrams with the sample locations and the sand fraction characteristics of principal components 1 and 2 only, depicted on the X- and Y-axes respectively. For a correct scaling, we used the focus on case distances and divide response variable scores by standard deviation.

For the coarse (250–2000  $\mu\text{m}$ ) sand fraction (Fig. 4A), principal component 1 explained 74% of the variance in geochemical composition and principal component 2 explained 12% of the variance. For both sites, samples taken in the inner dunes (zone 4) were clearly separated from the samples taken in the other zones for the first component, and this distribution correlated positively with C, N, exchangeable P, Ca-bound P, and organic P. For all zones, samples from SE were separated from samples from NWC for the second component, with SE samples being characterized by high contents of Fe oxides, aqua regia contents of Fe and K, and available P, and low contents of aqua regia Ca.

For the fine (<250  $\mu\text{m}$ ) sand fraction (Fig. 4B), the PCA results showed an explained variance of 56% for the first component and 30% for the second component. Again, zones 1–3 (on the left) were separated from zone 4 (on the right) for both locations, but the distinction was much less clear than for the coarse fraction, especially for samples from NWC. This indicates that the transportation of fine material was creating a geochemistry in the dune area that was more homogeneous than that of the coarse fraction. Samples from zones 1–3 of both SE and NWC were characterized by high Ca and Fe contents (except for Fe-bound P), with no clear distinction between zones and/or sample location. Interestingly, Fe-bound P was high for zone 4 samples from SE, whereas Fe oxides are low.

Fig. 4C presents the PCA of the fine (<250  $\mu\text{m}$ ) superficial sand samples with fine (<250  $\mu\text{m}$ ) sand trap samples of NWC only. The PCA showed an explained variation of 53% for the first component and 36% for the second component. Clearly visible from the figure was that all the sand trap samples were located in the lower right corner, together with superficial sand samples of zones 1 and 2. Therefore, the geochemical composition of trapped sand samples from zones 3 and 4 most closely resembled that of the superficial samples of zones 1 and 2. Both these superficial samples as well as trapped sand samples were characterized by high contents of aqua regia content of K, Ca-bound P, and – to a lesser extent – total P, detrital P, aqua regia contents of Fe and Ca, and other Fe characteristics. Superficial samples from zone 4 lay farther away from the trapped sand samples in Fig. 4C, indicating that their geochemical composition was less similar.

As a result, the coarse fraction showed the highest difference between zones 1–3 and zone 4 compared to the fine fraction. A clear distinction between the sites was shown as well, which was not visible using the three-way independent ANOVA. Both fractions showed that organic P, available P, exchangeable P and N had highest contents in zone 4, whereas Ca and Fe contents were highest in zone 1–3. Fe-bound P showed high contents for SE superficial samples and mainly in zone 4, whereas detrital P was seen at both sites with high contents for the fine fraction. The trapped sand resembled most closely the geochemical composition of superficial samples from zone 1 and 2 with Ca-bound P containing the highest content of the different P bindings.



**Fig. 4.** Ordination diagrams with the first two axes of a PCA of the chemical characteristics of the (A) coarse surface sand fraction, (B) fine surface sand fraction and the (C) fine fraction of trapped sand combined with the fine surface sand fraction. The dark blue arrows represent the different chemical elements where 'tot' stands for total, 'av' for available, 'ar' for aqua regia, 'Ox' for oxides, 'exP' for exchangeable P, 'FeP' for Fe-bound P, 'CaP' for Ca-bound P, 'detP' for detrital P. Each symbol represents a different site and each color a different zone.

### 3.4. Chemical characteristics of the plant material

Chemical characteristics for the two plant species *A. arenaria* and *C. arenaria* were listed in Table 5. A significant difference was found between SE and NWC for K and Ca in *C. arenaria*. No significant differences were found for *A. arenaria* between the two sites. Contents of P, Ca, and N were lower for *A. arenaria* compared to *C. arenaria*. Interestingly, *A. arenaria* at SE contained a much higher Fe content than *A. arenaria* at NWC, which might be related to the higher contents of Fe in the nourishment sand at SE.

For each site, significant differences were measured by comparing the four separate zones (Table 6). For SE, comparison of zones clearly revealed a higher Ca content in *C. arenaria* in zone 3 compared to zone 4, as well as a higher Fe content in *A. arenaria* in zone 1, with a decreasing trend toward zone 3 and a significant difference between zones 2 and 3. The same holds for K, where a decreasing trend was seen for *A. arenaria* in zones 1 to 4 at SE. The trend for Fe was not present at NWC, which

**Table 5**

Chemical characteristics of the vegetation at the Sand Engine (SE) and North West Core (NWC) with the means and standard deviations (st. dev). When the concentrations differ significantly for *C. arenaria* between NWC and SE (independent t-test,  $p < 0.05$ ), the variable is listed with a \*. No significant difference between SE and NWC found for *A. arenaria*.

|           | SE                          |                             | NWC                        |                            | Significance between NWC and SE |
|-----------|-----------------------------|-----------------------------|----------------------------|----------------------------|---------------------------------|
|           | <i>A. arenaria</i> (n = 20) | <i>C. arenaria</i> (n = 12) | <i>A. arenaria</i> (n = 8) | <i>C. arenaria</i> (n = 7) |                                 |
|           | Mean (st. dev)              | Mean (st. dev)              | Mean (st. dev)             | Mean (st. dev)             | <i>C. arenaria</i>              |
| Ca (mg/g) | 1.6 (0.5)                   | 2.5 (1.2)                   | 1.6 (0.6)                  | 3.7 (1)                    | *                               |
| Fe (mg/g) | 0.13 (0.2)                  | 0.06 (0.02)                 | 0.05 (0.03)                | 0.08 (0.02)                |                                 |
| K (mg/g)  | 5.1 (1.9)                   | 6.9 (1.7)                   | 5.3 (1.1)                  | 9.6 (1.2)                  | *                               |
| P (mg/g)  | 0.51 (0.2)                  | 0.86 (0.3)                  | 0.6 (0.16)                 | 1 (0.2)                    |                                 |
| N (mg/g)  | 6.2 (2.2)                   | 11 (3.4)                    | 7.5 (2.4)                  | 13 (3.4)                   |                                 |
| N:P       | 13.4 (5.4)                  | 15 (6.2)                    | 13 (4)                     | 14 (7)                     |                                 |
| N:K       | 1.3 (0.6)                   | 1.9 (0.8)                   | 1.5 (0.6)                  | 1.4 (0.4)                  |                                 |
| K:P       | 10.5 (2.9)                  | 8.5 (1.9)                   | 8.6 (1.4)                  | 9.6 (2.6)                  |                                 |

were also established for superficial sand with the PCA and the three-way independent ANOVA.

Additional statistical analyses were performed to test for differences between zones and between sites SE and NWC (independent t-test,  $p < 0.05$ ). In SE, *A. arenaria* had a significantly higher Fe content than in NWC. In zone 4, *C. arenaria* had a significantly lower content of Ca at SE than at NWC.

**4. Discussion**

*4.1. Characteristics of superficial sand at the beach and in the dune area*

At SE as well as NWC, the geochemical characteristics (including the binding of P) of the sand were found to vary with fraction (fine (<250 μm) versus coarse (250–2000 μm)) and zone (zones 1–3 versus the inland zone 4). The geochemical difference between fine and coarse sand is most likely attributable to variation in mineral composition, which results from preferential mineral transportation (Arens et al., 2002; Shao, 2009) in addition to hardness of minerals (Boggs, 2009).

The coarse fraction from the beach to the foredunes crest is dominated at both sites by two main P bindings: Fe-bound P and detrital P. Detrital P might include crystalline apatite, a mineral that has been observed in Dutch dune sands (Druif, 1927; Griffioen et al., 2016). The coarse fraction of the inner dunes, on the other hand, shows high contents of organic P, exchangeable P and, to a lesser extent, Ca-bound P, which might be related to the increase in organic matter as the C content of the material is higher in the inner dunes, especially at NWC where vegetated dune slacks are present in the hinter dunes.

The chemical composition of the fine fraction at NWC and SE was similar for the beach, the frontal foredunes and the foredunes crest as well. Both sites show high contents of Ca-bound P, which includes Ca-phosphates and CaCO<sub>3</sub>-associated P (Ruttenberg, 1992). Additionally, crystalline apatite or silicate-P seems to be present in these zones where detrital P, and total P are clustered together in the PCA. The contents of K and Fe in that same cluster may indicate the presence of heavy minerals as well. Heavy minerals are present at SE (Pit et al., 2017) and are also common in the wider Dutch coastal area (Eisma, 1968; Schüttenhelm and Laban, 2005). With a higher percentage of finer material at NWC compared to SE for the beach, frontal foredunes and foredunes crest, differences between the study sites are probably more apparent than estimated by measuring the two fractions separately.

At the inner dunes, the characteristics of the fine fraction do differ between the two sites. In this zone, P bound to Fe was higher at SE, whereas organic P and exchangeable P were higher at NWC. For SE, the high Fe-bound P content in the inner dunes and high Ca-bound P content at the beach, the frontal foredunes, and the foredunes crest create a familiar pattern for stable dune systems: Ca-bound P is assumed to be dominant in the seaward zone of the dune area and on the beach because of the high carbonate content (Lindsay and Moreno, 1960), whereas Fe is assumed to be more dominant further into the dune area, the inner dunes, because here a longer period of rainwater infiltration lowers pH and decalcifies the sand, causing Ca-bound P to dissolve (Kooijman et al., 1998). The presence of dune slacks in the inner dunes at NWC is probably giving rise to high contents of organic and exchangeable P. Organic matter builds up in large quantities in dune

**Table 6**

Chemical characteristics of the vegetation at Sand Engine (SE) and North West Core (NWC) per zone, with the means and standard deviations (st. dev). Zones that show a significant difference (Games Howell post hoc,  $p < 0.05$ ) between the other zones (within one site) are indicated with different letters. For *C. arenaria* at NWC, only one sample is present in zone 2 and therefore, an independent t-test was performed to establish significant differences.

|           |                    | SE           |              |               |                | NWC           |               |                |               |
|-----------|--------------------|--------------|--------------|---------------|----------------|---------------|---------------|----------------|---------------|
|           |                    | Zone 1       | Zone 2       | Zone 3        | Zone 4         | Zone 1        | Zone 2        | Zone 3         | Zone 4        |
| n         | <i>A. arenaria</i> | 4            | 8            | 4             | 4              | 2             | 4             | 2              | 0             |
|           | <i>C. arenaria</i> | 0            | 0            | 4             | 8              | 0             | 1             | 2              | 4             |
| Ca (mg/g) | <i>A. arenaria</i> | 1.8 (0.7) a  | 1.4 (0.4) a  | 1.9 (0.1) a   | 1.4 (0.4) a    | 1.6 (0.1) a   | 1.3 (0.4) a   | 2.3 (0.7) a    |               |
|           | <i>C. arenaria</i> |              |              | 3.4 (0.6) a   | 2.0 (1.2) b    |               | 3.9 (–) a     | 3.2 (0.3) a    | 3.9 (1.3) a   |
| Fe (mg/g) | <i>A. arenaria</i> | 0.3 (0.4) ab | 0.1 (0.04) b | 0.04 (0.01) a | 0.09 (0.11) ab | 0.08 (0.03) a | 0.03 (0.01) a | 0.06 (0.01) a  |               |
|           | <i>C. arenaria</i> |              |              | 0.06 (0.01) a | 0.06 (0.03) a  |               | 0.1 (–) a     | 0.07 (0.004) a | 0.08 (0.02) a |
| K (mg/g)  | <i>A. arenaria</i> | 6.4 (2.8) a  | 5.4 (1.3) a  | 4.9 (1.8) a   | 3.5 (1.1) a    | 5.8 (0.8) a   | 5.0 (1.2) a   | 5.2 (1.5) a    |               |
|           | <i>C. arenaria</i> |              |              | 7.6 (1.0) a   | 6.6 (1.9) a    |               | 9.3 (–) a     | 8.5 (0.9) a    | 10 (1) a      |
| P (mg/g)  | <i>A. arenaria</i> | 0.6 (0.4) a  | 0.5 (0.2) a  | 0.6 (0.1) a   | 0.4 (0.1) a    | 0.7 (0.1) a   | 0.5 (0.1) a   | 0.8 (0.3) a    |               |
|           | <i>C. arenaria</i> |              |              | 0.9 (0.3) a   | 0.8 (0.3) a    |               | 1.3 (–) b     | 1.0 (0.02) a   | 1.0 (0.2) ab  |
| N (mg/g)  | <i>A. arenaria</i> | 9.0 (3.5) a  | 4.9 (1.0) a  | 6.0 (0.5) a   | 6.4 (1.2) a    | 10 (3.3) a    | 7.1 (1.9) a   | 6.0 (0.4) a    |               |
|           | <i>C. arenaria</i> |              |              | 11 (3.1) a    | 12 (3.5) a     |               | 11 (–) a      | 13 (1.4) a     | 14 (4.5) a    |
| N:P       | <i>A. arenaria</i> | 18 (5.9) a   | 11 (5) a     | 11 (4.0) a    | 16 (4.8) a     | 14.7 (2.9) a  | 13 (4.1) a    | 8.5 (3.4) a    |               |
|           | <i>C. arenaria</i> |              |              | 12 (3.1) a    | 17 (6.9) a     |               | 8.8 (–) a     | 13 (1.7) a     | 15 (9.4) a    |
| N:K       | <i>A. arenaria</i> | 1.4 (0.3) a  | 0.9 (0.2) a  | 1.4 (0.7) a   | 2.0 (0.6) a    | 1.8 (0.8) a   | 1.5 (0.6) a   | 1.2 (0.5) a    |               |
|           | <i>C. arenaria</i> |              |              | 1.4 (0.6) a   | 2.1 (0.9) a    |               | 1.2 (–) a     | 1.5 (0.3) a    | 1.4 (0.5) a   |
| K:P       | <i>A. arenaria</i> | 12 (2.5) a   | 11.5 (3.1) a | 8.7 (1.8) a   | 8.4 (2.0) a    | 8.8 (2.3) a   | 9.2 (0.7) a   | 7.0 (0.02) a   |               |
|           | <i>C. arenaria</i> |              |              | 8.7 (2.3) a   | 8.4 (1.9) a    |               | 7.4 (–) a     | 8.8 (0.8) a    | 11 (3.1) a    |

slack marshes (Lammerts et al., 1999; Sival, 1996), whereas Fe oxides are less likely to be present due to reduction in anaerobic soils (Patrick and Khalid, 1974). The exceptionally high N content at the foredunes crest at NWC showed to be an outlier and was most likely influenced by bird droppings.

#### 4.2. Differences between the study sites

Comparison of the study sites revealed that Fe minerals are prevailing in the coarse fraction at SE and Ca minerals at NWC, despite the similarity in P bindings in zones 1–3 at both sites. Aside from the dune slacks present in the inner dunes of NWC, which result in high contents of organic-bound P, the differences between the two sites may be attributable to differences in source material, or the difference in age of the dune area.

SE contains material from two different geological epochs, the Holocene and the Pleistocene (Pit et al., 2017). In comparison with the Pleistocene material, the Holocene material contains more reactive Fe, which originates from the outflow of the River Rhine 10 km south of SE (Canavan et al., 2007). The beach at NWC, on the other hand, is nourished irregularly with a relatively low volume of sand and most recent sand nourishments are foreshore nourishments. Due to its low volume, it is likely that this sand comes from the uppermost, Holocene layer of the sand pit and has not been mixed with deeper, Pleistocene material (personal communication from Gert Jan Harpe, Ministry of Infrastructure and Water Management, the Netherlands). Additionally, the sand pit for this traditional nourishment is 50 km further north than SE and 60 km north of the Rhine outflow, and therefore received less reactive Fe than the sand pit of SE.

An age difference of the dune areas at the sites may have influenced the presence of certain minerals, too. Both dune areas are part of the so-called Holland coast, but NWC contains a wide range of Younger Dunes, originating from the Middle Ages, which are followed more inland by a range of old barrier dunes dating from 1000 CE (Doing, 1995). Because of their age, the old barrier dunes are strongly decalcified due to leaching (Eisma, 1968). In the dune area at SE, however, these two types of dune landscape are more or less mixed within the narrow dune range close to the beach. Therefore, decalcification is more likely to be visible at the inner dunes at SE, which might be one reason for the difference in pH (6.0 at SE versus 7.5 at NWC) and Ca content of the fine sand fraction ( $250 \text{ g m}^{-2} (5 \text{ cm})^{-1}$  at SE versus  $766 \text{ g m}^{-2} (5 \text{ cm})^{-1}$  at NWC). As both sites are located in carbonate-rich environment, this effect can become higher in the more northern regions of the Netherlands.

#### 4.3. Transportation of material from the beach toward the dune area

The coarse fraction is most frequently transported by rolling and saltation from the beach inland. During transport, the mean grain size and sorting decrease (Arens et al., 2002), which probably limits the transportation of the coarse fraction to the inner dunes. The fine fraction can be picked up by the wind and transported to the inner dunes. Our PCA results suggest that this is the case at NWC, where the geochemistry of the trapped sand from the dune area is similar to that of the superficial sand from the beach.

Trapped sand from NWC show that at least 80% of the eolian material was  $<250 \mu\text{m}$ , a median grain size distribution (D50) of 200–250  $\mu\text{m}$ , with a slightly lower D50 in the inner dunes, which was also found in a study by Van Buuren (2017). A comparable trend was visible in the grain size distribution at the surface of SE and NWC: the D50 was largest at the beach and smallest at the inner dunes. However, it seems that the nourishment material is finer at NWC than at SE with a much higher  $<250 \mu\text{m}$  fraction on the beach (41% versus 17%). Nourishment material is often coarser than the native beach material, the D50 of which has been found to be between 211 and 346  $\mu\text{m}$  (Van der Wal, 1999). With a D50 of 275  $\mu\text{m}$  at the beach, NWC falls within this

range, but SE contains coarser material with a D50 of 376  $\mu\text{m}$ . This may be related to differences in source material, as explained above.

As explained in 4.1, the fine fraction at NWC and SE showed a similar chemical composition for the beach, the frontal foredunes and the foredunes crest. For NWC, it seems that fine material from the beach is not transported beyond zone 3 at NWC, as also reported by Van Buuren (2017). It may take more time for the transport of fine material from the beach to develop, as the dynamic dune system has been created relatively recently (2012). In case of SE, this is not expected as this study site does not have dynamic foredunes.

For SE, the geochemistry remains the same from the beach to the foredunes crest, confirming our hypothesis that the presence of fixed foredunes would result in a similar geochemistry toward the dune area. However, we did expect that the foredunes crest would show a less similar geochemistry compared to the beach and the frontal foredunes. Therefore, it is plausible that enough fine material was transported to the dune area to create comparable superficial sand, although not to the inner dunes. A possible explanation for this is the nourishment history at SE where there have been 16 sand nourishments since 1986, 11 of them being beach or foredune replenishments with a total volume of almost  $34 \text{ M m}^3$  of sand (personal communication from Gert Jan Harpe, Ministry of Infrastructure and Water Management, the Netherlands). The area around NWC contains sand replenishments applied since 1990 and with a much smaller volume: seven sand replenishments were applied, five of them beach nourishments each comprising  $1.6 \text{ M m}^3$  of sand, which is less than one twentieth of the volume applied to the SE beach. Also, when SE was created, its design and location resulted in fine material being transported over a distance up to 3 km (Van der Valk et al., 2012), which is common in the first year after applying a beach nourishment (Van der Wal, 1999). As the volume of SE was much larger than a traditional nourishment, the amount of fine material in the first year might have been significant, even though the dune system was not dynamic.

#### 4.4. The availability of P and its relation to P speciation

At the inner dunes of both SE and NWC, the P content of the coarse fraction is dominated by CAEM-available P and total P, which are correlated to Fe-bound P according to the PCA. This indicates that P is adsorbed to Fe oxyhydroxides involved with organic complexes, creating a more labile P fraction that is easily available to plants (Kooijman et al., 2009).

For both sites, the total P of the fine fraction is highest at the foredunes crest, which is most likely related to eolian processes. In this zone, detrital P including crystalline apatite accounts for the largest P fraction of total P. However, the CAEM-available P content is highest at the inner dunes. Here, CAEM-available P is mostly connected to organic P and exchangeable P. Exchangeable P or loosely sorbed P is readily available for uptake, but for organic P to become available, mineralization has to occur (Zheng and Zhang, 2000). Microbes play a major role in transforming P, including decomposition with a release of CAEM-available P (Stevenson, 1986).

Between the sites it was expected that SE would show a lower CAEM-available P compared to NWC, because of a possible P speciation change to Fe-bound P which may be desorbed less easily compared Ca-bound P due to decalcification. However, both sites showed a high similarity in P bindings and P contents for zones 1–3, including CAEM-available P contents.

#### 4.5. The chemical characteristics of the plant species compared to those of the superficial sand

Some differences in the geochemistry of the superficial sand between SE and NWC were mirrored in the elemental composition of the plant material, which were mainly related to Ca and Fe characteristics as well as the K content. It was expected that P contents would differ

per plant species too, but this was not the case. We estimated the accuracy of the method of CAEM-available P by using the Spearman's correlation coefficient (Field, 2009). A positive correlation exists between CAEM-available P contents of the total sediment and the P contents of the plant species ( $R^2 = 0.575^2$ ,  $p = 0.002$ ), which means that 33% of the variation in P content in plants is accounted for by the P availability in the soil. Even though the CAEM method is accurate, neither of the two study sites did we observe different P bindings or different CAEM-available P contents between the different zones. Only the Fe content of *A. arenaria* showed interesting results. *A. arenaria* is known to grow in places with low nutrient availability and can be a dominant plant species at locations where Fe levels are low (Kooijman et al., 1998). However, high Fe contents are present in the superficial sand at SE, indicating that variables other than nutrient availability, such as sand burial, seem to be limiting growth and survival of *A. arenaria* (Nolet et al., 2018).

For N, we did observe a shortage for both plant species at SE and NWC, resulting in a lower N:P ratio in the frontal foredunes and the foredunes crest. Since ratios below 13.5 indicate N-limitation (Olde Venterink et al., 2003), it is safe to conclude that N is likely limiting plant growth in the frontal foredunes and the foredunes crest. This is most likely related to low organic matter in these zones (Kooijman and Besse, 2002), which is high at the inner dunes, especially at NWC because of the presence of dune slacks. For the beach, where high N:P ratios were found for *A. arenaria*, co-limitation of N and P (NWC) and P-limitation (SE) occurs probably steered by the calcareous conditions (Kooijman et al., 1998).

With a dynamic dune system, it can be expected that the sand fractions, especially the fraction < 250  $\mu\text{m}$ , will eventually be transported to the inner dunes with time, which will create a dominance of inorganic Fe and Ca minerals and as a result reduce availability of P. Consequently, sensitivity to N deposition is less than when phosphate is readily available (Kooijman et al., 2009). Additionally, plant species of dunes, many of them being endangered, may benefit from a P-limited environment (Wassen et al., 2005), which will occur when P is bound to Ca minerals or Fe oxyhydroxides – i.e., there is pH buffering of the soil – rather than to organic complexes (Bakker et al., 2005).

## 5. Conclusion

Our study shows that the geochemical characteristics of superficial sand in the dune area of the two study sites Sand Engine (SE) and North West Core (NWC) depend on the zone and sand fraction. Beach material is being transported by wind toward the dune area at both sites, but transport toward the inner dunes is limited. Total P content was comparable for each sand fraction at both sites and around 2 times higher in the superficial fine sand fraction compared to the superficial coarse sand fraction. An exception is the superficial coarse sand fraction at NWC, which shows influences of dune slacks at the inner dunes resulting in high contents of organic P and exchangeable P resulting in a high total P content. For the beach, the frontal foredunes and the foredunes crest, P was related to Fe and Ca at both study sites. Interestingly, P bound to Fe included both Fe oxides as well as organic complexes attached to Fe oxides. P bound to Ca includes Ca-phosphates and  $\text{CaCO}_3$ -associated P. The organic complexes are related to the CAEM-available P content, which were highest at the inner dunes for both sites and superficial sand fractions. Here, chemical pH-dependent processes may have played a role, causing P to adsorb to Fe oxides involved with organic complexes, which is a more labile P content. Both fractions in superficial sand showed a similar geochemistry from the beach toward the foredunes crest. For SE this was unexpected, since here, unlike in NWC, the dune system is not dynamic. However, the nourishment history of the SE area implies that a large volume of sand has been applied on the beach and foredunes, probably creating a temporary supply of fine material toward the dune area until the nourishment material had stabilized.

To conclude, the P speciation of the beach area and therefore nourishment material is different compared to the inner dune area, especially when dune slacks are involved. The P speciation alters the P availability, but is mainly related to the difference in organic matter and minerals and less with the difference of P bound to Ca or Fe minerals. The total P content, P bindings and CAEM-available P content between a mega sand nourishment and a traditional sand nourishment do not significantly differ, as well as the effect of a dynamic dune area, although time might be a crucial factor. This study was performed 4 years after the creation of a dynamic foredunes and we found no evidence that dominant dune plant species were influenced by changes in soil chemistry due to transport of nourishment material into the dunes. We expect that this will change over time, especially when a dynamic dune system is present, like at NWC, where the fine sand fraction can be transported further into the dune area, providing an enrichment of beach-related minerals which could reduce the availability of P. This may reduce the vegetation's vulnerability to N deposition and increase pH buffering of the soil.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2020.138233>.

## CRedit authorship contribution statement

**Iris R. Pit:** Formal analysis, Conceptualization, Investigation, Writing - original draft. **Martin J. Wassen:** Supervision, Conceptualization, Investigation. **Annemieke M. Kooijman:** Conceptualization, Formal analysis, Investigation. **Stefan C. Dekker:** Supervision, Writing - review & editing, Conceptualization. **Jasper Griffioen:** Writing - review & editing, Validation. **Sebastiaan M. Arens:** Investigation. **Jerry van Dijk:** Writing - review & editing, Validation.

## Declaration of competing interest

Potential conflicts do not exist.

## Acknowledgements

This work was supported by NatureCoast, a project of the Technology Foundation STW (12691) of the Netherlands Organization for Scientific Research (NWO). Joy Burrough provided a language edit of the final draft of the paper. We thank Gerard van Zijl for collecting sand trap samples at NWC and Dunea for collecting sand trap samples at the Sand Engine.

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