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Haloconduction as a remediation strategy: Capture and quantification of salts excreted by recretohalophytes

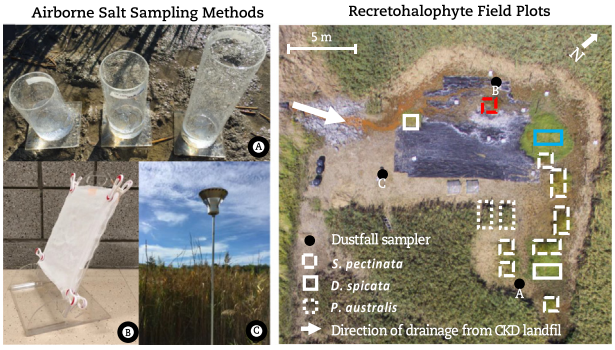
Kassandra B.M. Yun [a], Sonja Koster [b,*], Allison Rutter [a], Barbara A. Zeeb [a,b]

[a] School of Environmental Studies, Rm 0626 Biosciences Complex, Queen's University, 116 Barrie Street, Kingston, ON K7L 3N6, Canada
[b] Department of Chemistry and Chemical Engineering, Royal Military College of Canada, PO Box 17000, Station Forces, Kingston, ON K7K 7B4, Canada

HIGHLIGHTS

- Recretohalophytes *S. pectinata* and *D. spicata* are effective at remediating salinized soil through haloconduction
- Three methods developed to measure airborne salt excreted from recretohalophytes in the field
- Laboratory measurements of salt excretion rates indicate site-specific remediation timeframe of 1 – 3 years

GRAPHICAL ABSTRACT



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ABSTRACT

Recretohalophytes employ specialized glands to excrete salt ions onto their tissue surfaces, which then have the potential to be transported away from the plant via wind in a process referred to as 'haloconduction'. *Spartina pectinata* and *Distichlis spicata* were selected to investigate the potential to remediate a cement kiln dust landfill in Bath, ON via salt excretion and haloconduction. Under ideal conditions in the laboratory, measurements of salt excreted by large (>15 shoots and > 50 cm height) plants of each species were 280 ± 164 g/m^2 and 164 ± 75 g/m^2, respectively, resulting in potential remediation timeframes of 1.4 ± 0.9 and 2.4 ± 1.1 years. Three salt collection methods were developed and installed in the field to test their efficacy for capturing and measuring windborne salt mobilized from plant surfaces. All three methods (two ground-level and one at 260 cm height) were successful in capturing and quantifying airborne salts up to 15 m from the plots. This study is the first to collect and quantify dispersed salt from recretohalophytes and hence confirm the theory of haloconduction, a promising new remediation technology for salt-impacted soils.

* Corresponding author.
E-mail address: kosters@queensu.ca (S. Koster).

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1. Introduction

Soil salinization currently threatens ~7% of the world's land surface and is an expanding environmental problem caused by both natural and human activities (Hasanuzzaman et al., 2014; Li et al., 2014; Wakeel, 2013). Salinity affects plant function and health, and the health of aquatic communities (Li et al., 2014). The significant negative impact that salts have on agricultural production must also be addressed in order to mitigate soil degradation and the loss of agricultural land due to salinity.

In many cases of soil salinization, the main ion of concern is sodium (Na^+) due to its effect on soil structure, and its competition with potassium (K^+) in plant functions and uptake (Karadag et al., 2016; Wakeel, 2013). Chloride (Cl^-) toxicity is also associated with saline conditions, as Na^+ and Cl^- commonly occur in the soil together. At low concentrations, chloride is a plant micronutrient, but can be introduced into the soil environment in high, toxic quantities in different ways, including via leaching from cement kiln dust (CKD) landfills. CKD is a particulate waste by-product of the cement manufacturing process, with highly elevated concentrations of potassium chloride (KCl). The elevated levels of potassium allow the soil to maintain a high K^+/Na^+ ratio (McSorley et al., 2016a; McSorley et al., 2016b), resulting in chloride being the main ion of concern.

As an alternative to costly and resource-intensive excavations, chemical amendments, or flushing of the salinized soil, phytotechnologies using halophytes have been considered as low cost, in situ, and sustainable options for remediation (e.g. Devi et al., 2016; Farzi et al., 2017; Jesus et al., 2015; Muchate et al., 2016; Rozema et al., 2016;). Halophytes make up ~1% of the world's terrestrial flora and utilize a variety of mechanisms to tolerate high salinity environments (Flowers and Colmer, 2008). The mechanism employed by 'recretohalophytes' involves the use of specialized salt glands to excrete salts onto the plant's leaf surfaces in processes that are not yet fully understood (Dassanayake and Larkin, 2017). Yensen and Biel (2006) theorized that wind could continuously mobilize excreted salts from recretohalophytes into the air and move them away from a site of contamination to areas of lower salt concentration through a process they termed 'haloconduction.' Yensen and Biel (2006) estimate that in the case of *Distichlis*, 5–50 tons/ha/year of salt could be dispersed based on only 50% of the excreted salts becoming airborne. Preliminary research involving the recretohalophyte, *Distichlis spicata*, determined that soil characteristics were improved in a stand that had been established for eight years, compared to a stand where no *D. spicata* was established (Sargeant et al., 2008). Because salts are generally macro or micro nutrients at lower concentrations haloconduction has the potential to also improve surrounding soils as the salt is dispersed. The theory of haloconduction has not yet been proven in the literature, but it has the potential to be an environmentally-friendly, economical approach for the remediation of salt-contaminated soils, as salt could be continuously removed from the soil without intervention (i.e. plant harvest).

The aim of this study is to evaluate the validity of the theory of haloconduction, and the efficacy of its use as a remediation technique at an industrial cement plant where CKD was landfilled over a period of 30 years. Several studies have explored the structure and function of salt glands (e.g. Barhoumi et al., 2015; Ceccoli et al., 2015; Dassanayake and Larkin, 2017; Yuan et al., 2016), but salt excreted by recretohalophytes has never been measured under field conditions. Some promising preliminary results were shown by McSorley et al. (2016b), when they explored the potential for *Spartina pectinata* (prairie cordgrass) to remediate salinized soils in laboratory experiments using soil from the same site as this study. *S. pectinata* was therefore selected for further study, in addition to another recretohalophyte, *Distichlis spicata* (inland saltgrass). Sea salt aerosol collection methods were investigated and adapted to develop new collection methods appropriate for salts dispersed by recretohalophytes. The specific objectives were to: (i) quantify salt excretion by the two recretohalophytes in both a laboratory and field setting, (ii) develop salt collection methods to quantify the transport of windborne excreted salt in the field, and (iii) use the collected data to determine if haloconduction is occurring at the Lafarge CKD site in Bath, ON.

2. Materials and methods

2.1. Site description

The Lafarge Cement Manufacturing Plant is located in Bath, Ontario (76°48′ Long 44°10' Lat) in the Mixedwood Plains Ecozone, and is underlain by limestone. Its proximity to Lake Ontario contributes to consistent wind and precipitation from a lake effect throughout the year (Government of Canada, 2017). CKD was landfilled on the plant property from 1973 to 2003 at a rate of ~30,000 tons per year in unlined landfill cells of 27.9 ha in total. To the east of the landfill is a lower elevation marshland known as the 'cliff site' (~3000 m^2) which receives drainage from the CKD landfill, resulting in an area of ~1000 m^2 that is high in chloride (Cl^-) content (4730 ± 5980 μg/g; n = 100), with a sodium adsorption ratio (SAR) of 15.4, and was previously completely devoid of vegetation. Water from the site drains eastward into Bath Creek and then flows into Lake Ontario. In 2015 and 2016 experimental plots of *Spartina pectinata* and *Distichlis spicata* were established in the northeastern section of the site (Supplemental 1) following the success of preliminary laboratory work completed with *S. pectinata* (McSorley et al., 2016b).

2.2. Plant propagation

Plugs of *S. pectinata* seedlings (eight weeks old) were obtained from Norview Gardens Ltd. in Norwich, Ontario, and were transplanted (one plant per 10-cm pot) into high salinity (7000–16,000 μg Cl^-/g) CKD-contaminated soil from site in the greenhouse facility at the Royal Military College of Canada (RMC). Five kilograms of *D. spicata* seeds were donated by Brett Young, a seed production and distribution company based out of Calmar, Alberta. The seeds were germinated (by submerging seeds in a petri dish of tap water and leaving them undisturbed for 7–10 days), and then transplanted into potting soil and seedling trays. Once they were approximately 12 cm in height (~60 days), they were transplanted into high salinity CKD-contaminated soil from the site and were grown in the greenhouse facility at RMC. From September 2014 to September 2016, this process was repeated five times to expand the quantity and range of ages of both recretohalophyte species.

2.3. Quantification of salt excretion by recretohalophytes in the laboratory

In September 2016, three small (<5 shoots of ~15 cm in height), three medium (6–15 shoots of ~30 cm in height), and three large (>15 shoots of ~50 cm in height) plants of each species were selected from the greenhouse. These plants were placed on a plant stand with the sides covered by plastic wrap (to reduce physical disturbances) and grown under a 12 h fluorescent photoperiod for one week. Their conditions were maintained at an optimal temperature and humidity range from 15 to 25 °C and 55–65%, respectively (Morris et al., subm.). Plants were carefully removed one at a time to avoid disturbing any excreted and accumulated salt on their surfaces. They were then inverted into Ziploc bags and rinsed with deionized (DI) water to remove excreted salt. The volume of water used to rinse each plant was recorded such that the total mass of chloride collected could be calculated. Bags were sealed and stored in a fridge at 4 °C until analysis. Plants were not harvested following salt rinsing. Instead, plants of a similar size were harvested by cutting at the base of their shoots to determine mass. Based on these measurements, the following masses were assigned to the plant categories: *S. pectinata* – small = 0.6 ± 0.2 g (n = 8); medium = 7.2 ± 1.2 g (n = 8); large = 21.1 ± 4.8 g (n = 8); *D. spicata* – small = 0.4 ± 0.1 g (n = 6); medium = 3.6 ± 0.7 g (n = 6); large = 10.5 ± 1.9 g (n = 6) (Supplemental 2).

Analysis was completed at the Analytical Services Unit (ASU) at Queen's University. A subsample was filtered through a 0.45 μm filter prior to analysis by ion chromatography (IC) with a Dionex HPLC (High Performance Liquid Chromatography) system (ICS 3000), using an AG4A-SC guard column, an AS4A-SC analytical column, a carbonate/bicarbonate eluent, and a conductivity detector. The column flow rate was set to 2.0 mL/min. Prior to analysis, conductivity was measured.

The following calculation was used to determine the mass of salt (Cl^-) that could be theoretically extracted from the site by the recretohalophytes (adapted from McSorley et al., 2016a):

Mass of chloride removed per harvest (kg)
= (Cl Excretion × Time × Biomass × Area)/Conversion Factor

Cl Excretion = mean chloride concentration excreted per gram of plant in one week ($\mu g_{Cl-}/g$).
Time = number of weeks plants are excreting in one year.
Biomass = maximum shoot biomass per square meter (from literature) (g/m^2).
Area = total chloride-contaminated area (1175 m^2).

Conversion Factor = 1.0×10^9 (to convert from μg to kg).

2.4. Quantification of salt excretion by recretohalophytes in the field

To allow excreted salts to accumulate on the recretohalophytes undisturbed in the field, miniature greenhouses (122 cm × 122 cm × 88 cm) were obtained and installed simultaneously over one in situ plot of *D. spicata* and one of *S. pectinata* from May 26, 2017 – September 15, 2017. Greenhouse vents were opened and closed strategically to moderate the inside temperatures. Early in the season (May 2017), shoots of *S. pectinata* and *D. spicata* were ~30 cm and ~15 cm in height, respectively, while later in the season (Aug. 2017), they were ~100 cm and ~40 cm in height, respectively. Each week, triplicate shoots were carefully cut at their base, placed in a labelled Whirl-Pak® bag, and transported to the RMC laboratory.

Weekly temperature (°C), wind speed (m/s) and wind direction (°) data were obtained from a nearby (~300 m) weather station (Government of Canada, 2017). Due to seasonally higher than normal levels of precipitation, high humidity, and wet/flooded conditions at the site in 2017, some trials were omitted from collection. The samples that were included in analysis were collected on June 1 and 22, July 6 and 20, August 10 and 31, and September 7, for a total of seven trials.

Each sample bag had 15 mL of DI water added to it before it was shaken to thoroughly rinse the salt from the shoot surfaces. This was repeated in triplicate for a total rinse volume of 45 mL. The rinse water samples were stored in the fridge until analysis, filtered and prepared for analysis as described in Section 2.3.

2.5. Airborne salt collection methods in the field

In order to investigate the transport of excreted salts from plants in the field, three collection methods: i) columns, ii) cheesecloth mounts,

Fig. 1. A) Collection columns, 17.8, 25.4, and 35.6 cm in height. B) Cheesecloth mount (24 × 24 cm) with all-plastic clothespins. C) Environment Canada dustfall sampler.

and iii) dustfall samplers were designed, constructed, deployed, and assessed for their ability to collect windborne salt.

2.5.1. Columns

Acrylic pipe with a diameter of four inches was cut into various heights (17.8, 25.4, and 35.6 cm). These were then secured to a small, square base of cast acrylic sheet to create collection columns (Fig. 1A). The columns were filled with 250 mL DI water and deployed for a period of one week at a distance of 1 m from the four edges of a 1 m^2 plot of *S. pectinata* (NE, NW, SE, SW) with a column of each of the three heights on each side. The amount of water in each of the columns at the end of the trial was highly variable depending on the amount of precipitation/evaporation that occurred. The water in each of the columns was poured into a 500-mL container and each column was rinsed three times with a total of 50 mL DI water. This experiment was repeated for a total of four trials. The sample volume in the columns was recorded. Subsamples were analyzed as described in Section 3.3. This same method was completed for samples collected in Sections 2.5.2 and 2.5.3.

2.5.2. Cheesecloth mounts

Prior to the deployment of cheesecloth mounts, the site was covered with a landscape fabric barrier to limit the influence of soil particles on the samples collected. Acrylic stands were constructed to hold sheets of cheesecloth at an angle of ~60° (Fig. 1B). This method was adapted from Lomas and Gat (1967), who collected and measured the impact of sea salt aerosol on a coastal orange grove in Israel. Cheesecloth (80 grade, threadcount = 40 × 32 threads/in.) was purchased from Nusso Textiles in Toronto, Ontario, and cut into 576 cm^2 (24 × 24 cm) sheets. Two sheets of cheesecloth were secured using all-plastic clothespins. This allowed the window of cheesecloth to capture any airborne salt from the plots. Four cheesecloth mounts were placed at ground-level 1 m from the edge of each plot in all four directions (NW, NE, SW, and SE), and left for a period of one week. Samples were collected by carefully removing the cheesecloth without disturbing the accumulated salt, folding it in on itself and placing it in labelled Whirl-Pak® bags. All samples were stored in a fridge until further processing. This experiment was repeated in triplicate for plots of both *S. pectinata* and *D. spicata*, and at a distance of two and three meters away from the edge of each plot for a total of 81 samples (Supplemental 3).

Each cheesecloth sample was carefully placed into a 1-L Mason jar. The Whirl Pak® bag was rinsed in triplicate with 20 mL DI water and this water was poured into the Mason jar with the cheesecloth. An additional 40 mL was added to the Mason jar for a total volume of 100 mL DI water. The jar was sealed and then manually shaken for 1 min. The rinse water was then poured into a collection vessel and all water was extracted from the cheesecloth. The cheesecloth was rinsed two more times with 100 mL DI water, shaken for 1 min and all aliquots were combined. Cheesecloth controls were placed in the same locations as the column controls and all had <1000 μg of salt. Clean sheets of cheesecloth were sampled and all had <325 μg of salt.

2.5.3. Dustfall samplers

Three dustfall samplers were obtained from Environment Canada (American Society for Testing Materials, 2010) (Fig. 1C), and placed on metal poles, positioned 260 cm above the ground at locations 3, 10, and 15 m from the plots (Supplemental 1). Plastic bags filled with 500 mL DI water were secured in the dustfall samplers and were left for ~30 days at a time according to the standardized method (American Society for Testing Materials, 2010). To sample, the bags were carefully removed, sealed, and transported back to RMC. Dustfall sampling was repeated over the course of two field seasons a total of nine times (n = 27 per sample). Field blanks were obtained by setting each sampler in position for one minute and then taking them down for analysis. Dustfall field blanks for samplers A, B, and C were measured at 60 ± 30 μg, 55 ± 35 μg, and 55 ± 30 μg respectively (n = 5).

2.6. Quality assurance & quality control

One method blank and one Environment Canada certified reference material (CRM), Cranberry-05, were included for each batch of samples that were analyzed by inductively coupled plasma optical emission spectroscopy (ICP-OES). One analytical duplicate and internal QC samples were also completed for every 10 samples analyzed. The mean relative standard deviation for the duplicate samples were all <2%. All of the blanks were below detection limits and the quality control standard was within 5% of the target. The chloride concentration for the environment Canada CRM Cranberry-05 was within 10% of the certified value for all analyses.

2.7. Statistical analysis

Statistical analysis of the data was performed using Prism 7 for Mac OS X version 7.0d (GraphPad Software, Inc., USA) and SPSS (Statistical Package for the Social Sciences) Statistics Version 24 (IBM Analytics, NY, USA). All data was first tested for normality using the Shapiro-Wilk test. Before a two-way analysis of variance (ANOVA) was performed, the homogeneity of variances was tested with Levene's test. This analysis was then followed by a post hoc Tukey comparison. Multiple regression and two-way analyses of covariance (ANCOVA) were performed for those samples that were potentially affected by weather covariates. Principal component analyses were completed using the XLSTAT add-in by Addinsoft for Microsoft Excel.

3. Results and discussion

3.1. Salt excretion by recretohalophytes in the laboratory

Under optimal conditions (i.e. 15–25 °C and 55–65% humidity; Morris et al., subm.), the amount of salt excreted by the large *S. pectinata* plants was significantly higher than all other plant and size categories ($p < 0.05$) (Fig. 2).

The mass of chloride excreted per gram of plant tissue over one week was calculated (Table 1). For both plant species, the larger the plant, the more efficient they were at excreting salt, although this was only significant for *S. pectinata* ($p < 0.05$). The values reported for both plant species are higher than those reported by McSorley et al. (2016b), whose plants had a lower mean mass.

To account for variable conditions in the field, the conservative assumption that half of the growing season was appropriate for salt excretion was adopted (McSorley et al., 2016b). Hence, the phytoextraction potential of *S. pectinata* and *D. spicata* over an eight-week period (based on a 16-week growing season) was calculated (Table 1). The

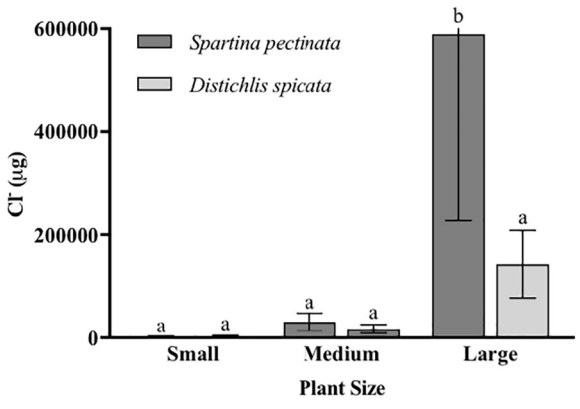

Fig. 2. Mass of chloride (μg) in water after rinsing stem and leaf surfaces of small (n = 3), medium (n = 3), and large (n = 3) *S. pectinata* and *D. spicata* plants after one week of salt accumulation. Error bars represent one standard deviation and significant differences are represented by lowercase letters.

Table 1
Mean weekly chloride (Cl^-) excretion on the stem and leaf surfaces of *S. pectinata* (n = 18) and *D. spicata* (n = 24) per gram of plant mass. Comparison of the phytoexcretion potential (g/m²), phytoexcretion rate (kg_{Cl-}), and remediation timeframe (yrs) of different size classes of *S. pectinata* and *D. spicata* in the laboratory over a period of eight weeks.

S. pectinata

Size	Excretion per plant ($\times 10^3$) (μg/g) dry weight per week	Phytoexcretion potential over 8 weeks (g/m^2)[1]	Salt removed per season (kg_{Cl-})	Remediation timeframe (yrs)
Small	3.4 ± 2.2^{A}	41.6 ± 26.5^{a}	48.8 ± 31.2	9.5 ± 6.1
Medium	4.3 ± 2.4^{AB}	52.4 ± 28.4^{a}	61.6 ± 33.4	7.5 ± 4.1
Large	22.8 ± 13.6^{B}	280.0 ± 164^{b}	324.0 ± 193.0	1.4 ± 0.9

D. spicata

Size	Excretion per plant ($\times 10^3$) (μg/g) dry weight per week	Phytoexcretion potential over 8 weeks (g/m^2)[1]	Salt removed per season (kg_{Cl-})	Remediation timeframe (yrs)
Small	7.1 ± 4.6^{AB}	85.2 ± 55.9^{ab}	100 ± 65.6	4.6 ± 3.0
Medium	8.2 ± 4.3^{AB}	98.9 ± 52.2^{ab}	116.2 ± 61.3	4.0 ± 2.1
Large	13.6 ± 6.2^{AB}	164.0 ± 75.0^{ab}	193.0 ± 88.1	2.4 ± 1.1

Uppercase letters indicate significant differences between the salt excretion per plant dry weight per week between *S. pectinata* and *D. spicata* ($p < 0.05$). These values were calculated based on mean biomass for each size category. Lowercase letters indicate significant differences between the phytoexcretion potential of *S. pectinata* and *D. spicata* over 8 weeks ($p < 0.05$).
[1] Phytoexcretion potential values (g/m^2) were calculated based on the plant maximum potential biomass in the literature (*S. pectinata* = 1510 g/m^2 (Helios et al., 2014); *D. spicata* = 908 g/m^2 (USDA, 2018).

total chloride in the top 10 cm of the soil profile at the CKD site in Bath, ON was estimated to be 395 ± 165 g/m^2 (McSorley et al., 2016b). To determine the feasibility of using halophytes and haloconduction to remediate the site, the time required to achieve remediation was calculated. Typical phytoremediation timeframes in the literature can sometimes require decades (Chen et al., 2015). We calculated that even the small plants of both *S. pectinata* and *D. spicata* would require less than one decade to remediate the site under ideal conditions, and that the large plants would require only 1.4 and 2.4 years, respectively (Table 1). This is consistent with McSorley et al. (2016b), who stated that the phytoremediation timeframe for this site via *S. pectinata* was ~3.5 years (using smaller plants ~20–30 cm in size). One of the challenges with phytoremediation is the long time frame – often at least five years to reach maturity and with lifespans of 50+ years (Kennen and Kirkwood, 2015) - and hence this is a very promising result. The reduced timeframe in combination with no requirement for plant harvest indicates that haloconduction will be a very cost effective phytotechnology.

3.2. Salt excretion by recretohalophytes in the field

Having successfully established salt excretion rates in the laboratory, similar experiments were carried out in the field on plots that had been established in previous years. Seven trials were carried out, and in each case, the ability of the plants to excrete salt onto their stem and leaf surfaces was confirmed (Supplemental 4). Plant sizes in June were comparable to the 'medium-sized' category of laboratory plants, and all other plants were comparable to, or larger than, the 'large' laboratory plants. The plant species' ability to phytoextract and excrete salt onto their stem and leaf surfaces was not significantly different ($p > 0.05$), with the exception of the trial completed on July 06, 2017 ($p < 0.05$). The chloride excreted by single *S. pectinata* or *D. spicata* plants varied widely, likely due to the high degree of meteorological variability and the adverse effects (high humidity) of the miniature greenhouses.

3.3. Airborne salt collection methods in the field

3.3.1. Columns

Columns (Fig. 1A) deployed 1 m from a 1 m^2 *S. pectinata* plot successfully captured airborne salt in the field (Fig. 3).

A multiple linear regression was employed to determine that the height (of the columns) and wind speed significantly contributed to the mass of chloride collected in the columns ($p < 0.05$), and that 49% of the variance in [Cl^-] was accounted for by the model ($F_{5,42} = 8.1$, $p < 0.05$). Column height had a negative influence on the amount of chloride collected, indicating that the higher the column, the less salt it collected. This is supported by PCA analysis (Fig. 4). Wind speed also correlated negatively, indicating that as wind speeds increased, less salt was collected by the columns. At higher wind speeds, the salt is potentially travelling further than the columns (placed at a distance of 1 m from the plot) before settling. This result is promising, as it indicates that salt may be travelling beyond the measured distance. Smaller and therefore more buoyant salt particles may travel hundreds of kilometers via wind transport before sedimenting (Morcillo et al., 2000). Over the course of four trials, the average mass of salt measured in the columns (without considering their height) was greater in the NW (1650 ± 2470 μg) and NE (1170 ± 1540 μg) directions than in the SW (991 ± 962 μg) and SE (1100 ± 927 μg) directions. These differences were not statistically significant ($p > 0.05$), however the prevailing winds are from the north, further supporting the theory that the salt is windborne. Nevertheless, as wind direction was found to be non-significant, chloride data for all directions was averaged for each trial and column height prior to PCA analysis. Trials 1 and 2 were completed in June and July while Trials 3 and 4 were completed during the month of October. The data points for Trials 1 and 2 appear on the right-hand side of the plot (Fig. 4) which corresponds with higher temperatures, while Trials 3 and 4 correspond with lower temperatures. Hence, time

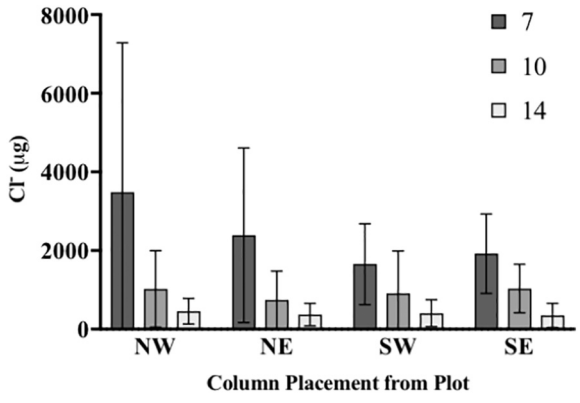

Fig. 3. Mass of chloride (μg) measured in water of columns of varying heights (17.8, 25.4, & 35.6 cm) placed 1 m from the edges of a plot of *S. pectinata* in four directions following seven days of collection (n = 48). Error bars represent one standard deviation. Each experiment included one set of columns (i.e. one column each of 17.8, 25.4, and 35.6 cm) placed 6 m from the edge of the plot in the SW direction and 9 m in the SE direction to act as controls, and all these controls collected <200 μg of salt (data not shown).

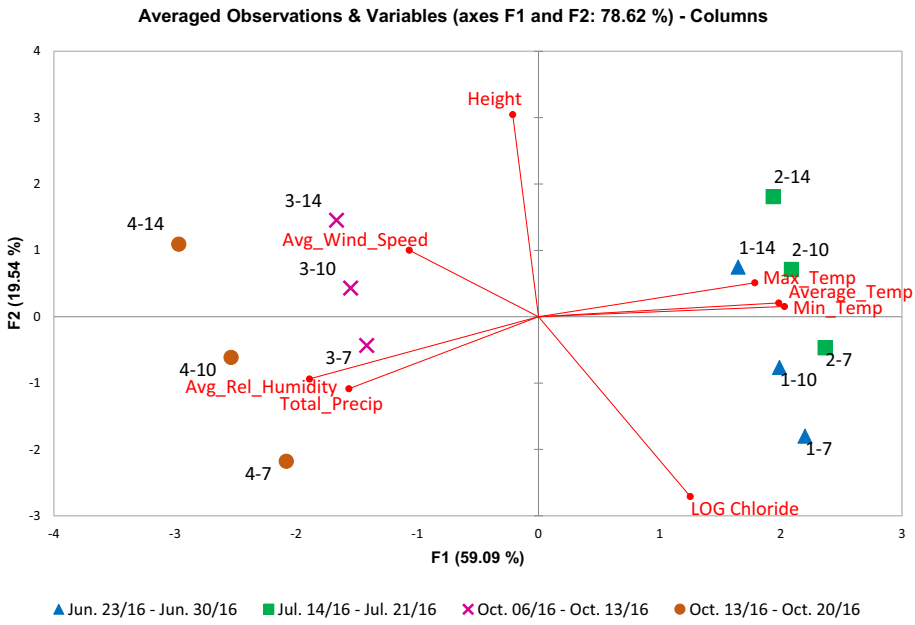

Fig. 4. Principal component analysis of column sampler data. Data point labels denote trial number and column height (Trial #_Height) and are grouped by sampling time range (Trial #1 = June 2016; Trial #2 = July 2016; Trials #3 and #4 = October 2016).

of year (correlating with temperature) is clearly an important indicator of salt excretion and subsequent dispersal.

3.3.2. Cheesecloth mounts

The cheesecloth mounts placed 1 m from the edges of the plots collected a higher mass of salt (5050 ± 4370 μg, n = 24) than the ones at 2 m (1590 ± 855 μg, n = 24) and 3 m (928 ± 843 μg, n = 24) (Fig. 5). PCA analysis (Fig. 6) clearly illustrates that the amount of chloride captured is negatively correlated to distance from the plot.

A multiple linear regression was used to determine that 52% of the variance in chloride could be accounted for using measured meteorological variables ($F_{8,63} = 5.8$, $p < 0.05$). Humidity had a significant ($p < 0.05$) negative influence on the amount of chloride, indicating the higher the humidity, the lower the amount of salt collected on the cheesecloth. This is consistent with the laboratory excretion observations. The wind direction again showed a general, but non-significant trend with higher amounts of salt collected in the NE and NW, suggesting that the salts are predominantly dispersed in the direction of the prevailing wind direction.

The chloride data for all directions was averaged for each trial prior to PCA analysis (Fig. 6). Sampling trials which took place later in the season appear on the left-hand side of the plot which is associated with lower temperatures. Chloride is negatively correlated with distance from the plot, and some grouping is apparent with the 1 m trials having highest chloride and 3 m trials having lowest chloride values. However, since all 3 m cheesecloth trials occurred later in the season it may be

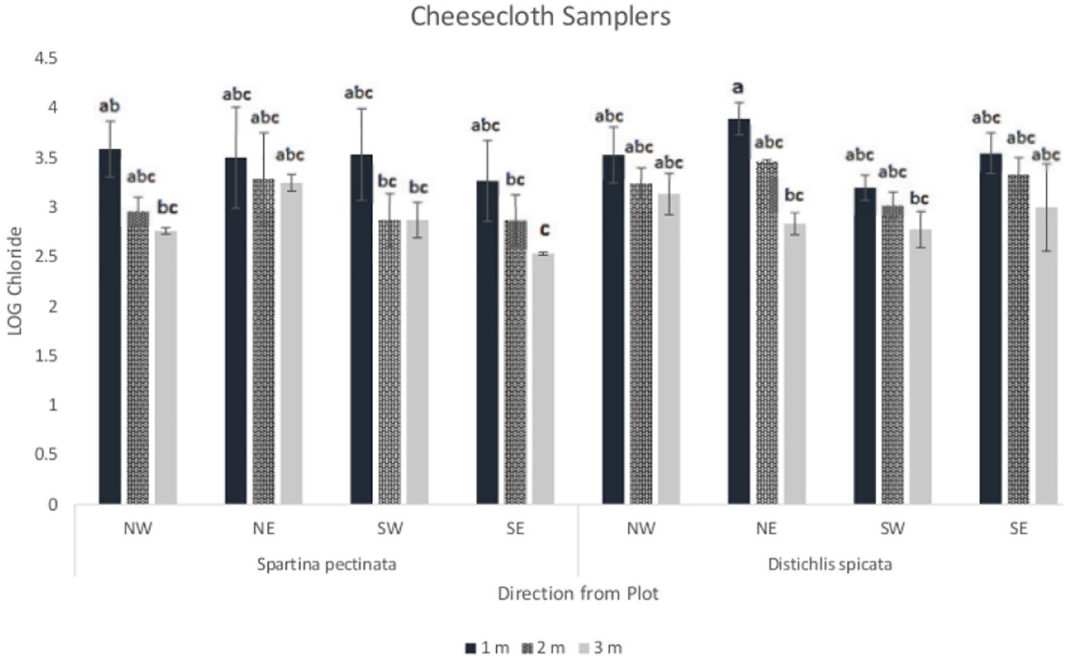

Fig. 5. Mass of chloride (μg) measured from rinsed cheesecloths that were placed on mounts at distances of 1, 2, and 3 m from the edges of individual plots of *S. pectinata* and *D. spicata* in all four directions (NW, NE, SW, and SE) (n = 72). Error bars represent one standard deviation and significant differences are represented by lowercase letters.

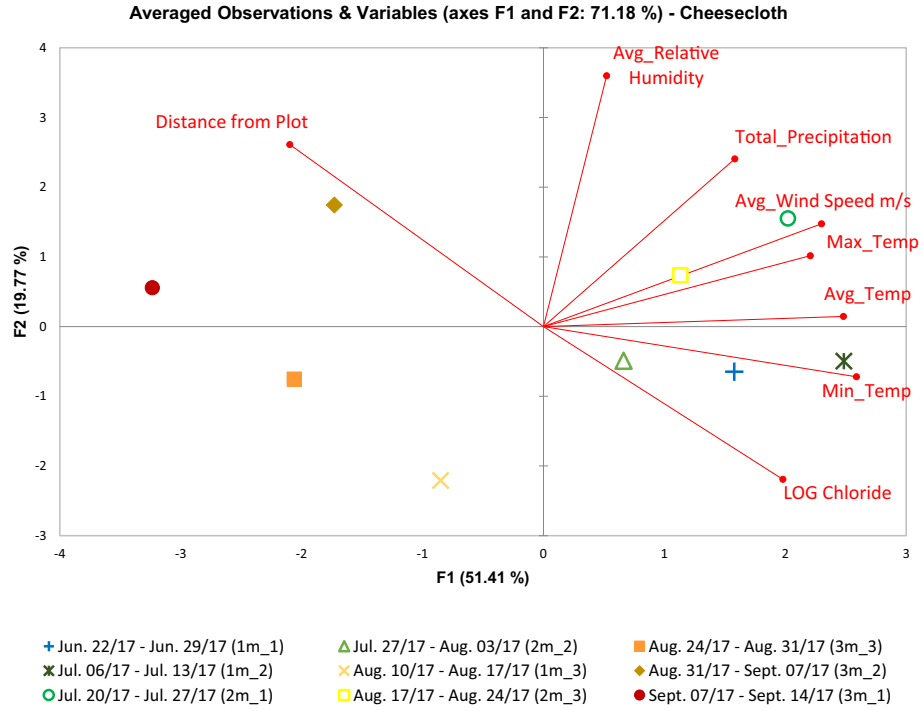

Fig. 6. Principal component analysis of cheesecloth sampler data. Data points are coded according to sampling time range and whether sampling occurred in early, mid, or late growing season. Values in brackets denote sampler distance and trial number (Distance (m)_Trial #).

difficult to differentiate the effects of temperature from the effects of distance.

To estimate losses due to precipitation, several cheesecloth trials were placed in trays such that any salt washed off during a rainstorm could be conserved. A mean of 1050 ± 341 μg/mL of salt was retrieved from the trays during any one trial (data not shown), increasing the total amount by 41.8 ± 13.6% (n = 27). Hence, the total amount of salt collected for *S. pectinata* at 1 m is potentially 7860 ± 7400 μm. In future trials, it will be important to improve the cheesecloth mounts (e.g. a small roof to shelter the cheesecloth from rain, similar to methods created by Gustafsson and Blomqvist, 2005) to include the collection and quantification of salts that are washed from off of the cheesecloth between sampling events. Field methods, particularly when sampling occurs over a long timeframe, are inherently variable given the wide variations in climate and plants. While it was imperative to trial haloconduction in the field, controlled laboratory experiments using a wind tunnel are currently underway and will allow improved characterization of the potential of haloconduction.

3.3.3. Dustfall samplers

Dustfall samplers (Fig. 1C) collected lower levels of salt in comparison to the cheesecloth mounts and columns, likely due to their positioning further from the plots and at higher elevations. However, these results provide further evidence of haloconduction, as they confirm that salt is being collected and measured at further distances from the plots and at locations higher than ground-level. There was no statistical significance between the amount of chloride found in dustfall samplers located at the different positions of A, B, and C (Supplemental 1). There were some temporal differences, in that the trial during September–October 2017 was significantly higher in mass of chloride (1770 ± 377 μg) than all other trials aside from June–July 2016 (1000 ± 312 μg) and May–June 2017 (1120 ± 708 μg) ($p < 0.05$) (Fig. 7). The lowest mass of chloride measured in the dustfall samplers was in June–July 2017 (215 ± 55 μg). Overall, dustfall samplers A, B, and C had a mean mass of chloride of 799 ± 679 μg, 868 ± 514 μg, and 568 ± 483 μg, respectively (n = 9).

A multiple linear regression was used to determine that wind speed significantly contributed to the chloride found in the dustfall samplers ($p < 0.05$) and accounted for 37% of the variance ($F_{7,19} = 1.59$, $p > 0.05$). PCA analysis indicates the data points show distinctive clustering by time of sampling, but not by sampler location (Fig. 8). The dustfall data points appear less distinctly effected by temperature than the other samplers and chloride is negatively correlated to precipitation. The lack of the distinct seasonal grouping seen in the ground level samplers could indicate that there is more variation in salt distribution patterns at greater distances, or that these particular meteorological factors have less dramatic effects at greater distances.

Salt is deposited into the columns and dustfall samplers as salt particles drop out of the atmosphere. The successful collection of salt via these two methods proves that the salt is travelling via haloconduction

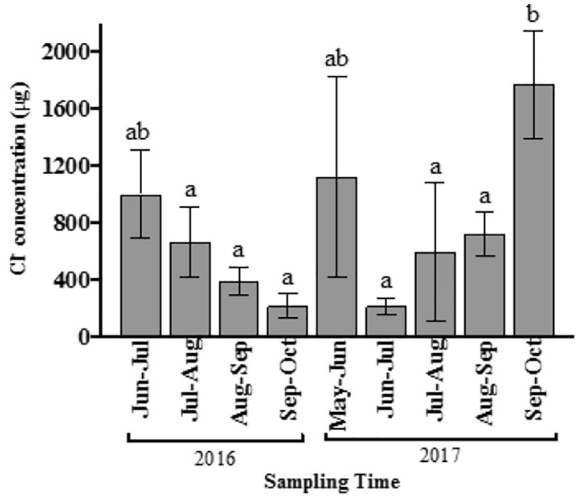

Fig. 7. Mean monthly mass of chloride (μg) obtained via dustfall samplers at the CKD site during the 2016 and 2017 field seasons. Error bars represent one standard deviation and significant differences are represented by lowercase letters.

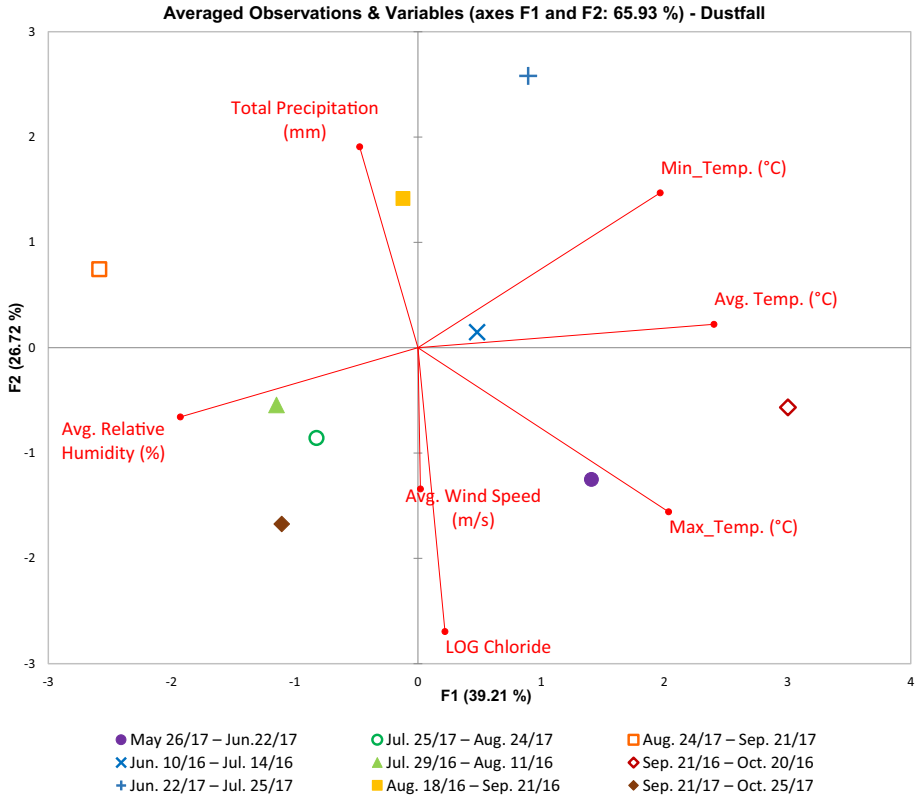

Fig. 8. Principal component analysis of dustfall sampler data. Data points are coded according to sampling time range and whether sampling occurred in early, mid, or late growing season. The relative position of each sampler to the plot was found to be non-significant, thus the chloride data for all three dustfall samplers was averaged for each trial prior to PCA analysis.

and is being deposited further distances away from the source of contamination.

3.3.4. Comparison of methods

In order to compare the two ground-level collection methods, the mean of the three highest results for each method at 1 m were calculated and determined to be 5750 ± 3030 μg and 13,200 ± 3230 μg chloride for column (10 cm width) and cheesecloth (24 cm width), respectively. The consistency of results from the two ground-level collection methods (columns and cheesecloth mounts), confirms the efficacy of these methods in capturing airborne salt. It is interesting to note that the columns, which require settling of airborne salt particles, are collecting similar amounts of salt as the cheesecloth mounts.

Combined PCA analysis of all sampling methods displayed a relatively clear grouping of mid-late season (and May–June) data points corresponding to lower temperatures and early-mid season data points corresponding to higher temperatures. Chloride was negatively correlated to average wind speed and total precipitation (Supplemental 5).

This study was the first to demonstrate the theory of haloconduction in the field. Smaller and therefore more buoyant salt particles are expected to have travelled further than the points of measurement in this study, whereas larger particles are expected to have been collected due to their lower suspension time in the atmosphere. While this study focused on KCl, studies currently underway with NaCl are expected to show similar results. Our results are consistent with the postulation of Yensen and Biel (2006) and the preliminary research with *Distichlis spicata* by Sargeant et al., 2008). It is now possible to apply the methods developed in this study to additional CKD, and other types of salt-impacted, sites. This research provides promising results to further develop the use of recretohalophytes and the theory of haloconduction as a phytoremediation technique for salt-impacted soils.

Declaration of Competing Interest

The authors declare no competing financial interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.06.271.

References

American Society for Testing and Materials, 2010. Standard Test Method for Collection and Measurement of Dustfall (Settleable Particulate Matter), D1739-98. ASTM International, West Conshohocken, PA.

Barhoumi, Z., Atia, A., Trabelsi, N., Djebali, W., Chaïbi, W., Ben Ammar, A., Abdelly, C., Smaoui, A., 2015. Scanning and transmission electron microscopy and X-ray analysis of leaf salt glands of *Limoniastrum guyonianum* Boiss. Under NaCl salinity. Micron 78, 1–9.

Ceccoli, G., Ramos, J., Pilatti, V., Dellaferrera, I., Tivano, J.C., Taleisnik, E., Vegetti, A.C., 2015. Salt glands in the Poaccae family and their relationship to salinity tolerance. Bot. Rev. 81, 162–178.

Chen, J., Chen, Y., Shi, Z., Su, Y., Han, F.X., 2015. Phytoremediation to remove metals/metalloids from soils. In: Ansari, A.A., Gill, S.S., Gill, R., Lanza, G., Newman, L. (Eds.), Phytoremediation: Management of Environmental Contaminants. vol. Volume 2. Springer International Publishing Switzerland, Switzerland, pp. 297–304.

Dassanayake, M., Larkin, J.C., 2017. Making plants break a sweat: the structure, function, and evolution of plant salt glands. Front. Plant Sci. 8, 1–20.

Devi, S., Nandwal, A., Angrish, R., Arya, S., Kumar, N., Sharma, S., 2016. Phytoremediation potential of some halophytic species for soil salinity. Int. J. Phytoremediation 18 (7), 693–696.
Farzi, A., Borghei, S.M., Vossoughi, M., 2017. The use of halophytic plants for salt phytoremediation in constructed wetlands. Int. J. Phytoremediation 19 (7), 643–650.
Flowers, T.J., Colmer, T.D., 2008. Salinity tolerance in halophytes. New Phytol. 179, 945–963.
Government of Canada, 2017. Historical climate data. Retrieved from. http://climate.weather.gc.ca/.
Gustafsson, M., Blomqvist, G., 2005. Modeling Exposure of Roadside Environment to Airborne Salt: Case Study. From Transportation Research Circular E-C063. Transportation Research Board, National Research Council, Washington, D.C., pp. 296–306 Retrieved from. https://www.vti.se/en/Publications/Publication/modeling-the-exposure-of-roadside-environment-to-a_670267.
Hasanuzzaman, M., Nahar, K., Alam, M.M., Bhowmik, P.C., Hossain, M.A., Rahman, M.M., ... Fujita, M., 2014. Review article: potential use of halophytes to remediate saline soils. Biomed. Res. Int. 2014 (589341), 1–12 Hindawi.
Helios, W., Kozak, M., Malarz, W., Kotecki, A., 2014. Effect of sewage sludge application on the growth, yield and chemical composition of prairie cordgrass (*Spartina pectinata* Link.). J. Elem. 19, 1021–1036.
Jesus, J., Danko, A., Fiuza, A., Borges, M., 2015. Phytoremediation of salt-affected soils: a review of processes, applicability, and the impact of climate change. Environ. Sci. Pollut. Res. 22, 6511–6525.
Karadag, S., Eren, E., Cetinkaya, E., Özen, S., Deveci, S., 2016. Optimization of sodium extraction from soil by using a central composite design (CCD) and determination of soil sodium content by ion selective electrodes. Eurasian J. Soil Sci. 5 (2), 89–96.
Kennen, K., Kirkwood, N., 2015. Phyto: Principles and Resources for Site Remediation and Landscape Design. Routledge Taylor & Francis Group, London and New York (346 pp).
Li, J., Pu, L., Han, M., Zhu, M., Zhang, R., Xiang, Y., 2014. Soil salinization research in China: advances and prospects. J. Geogr. Sci. 24 (5), 943–960.
Lomas, J., Gat, Z., 1967. The effect of windborne salt on citrus production near the sea in Israel. Agric. Meteorol. 4, 415–425.
McSorley, K., Rutter, A., Cumming, R., Zeeb, B.A., 2016a. Phytoextraction of chloride from a cement kiln dust (CKD) contaminated landfill with *Phragmites australis*. Waste Manag. 51, 111–118.
McSorley, K.A., Rutter, A., Cumming, R., Zeeb, B.A., 2016b. Chloride accumulation vs chloride excretion: phytoextraction potential of three halophytic grass species growing in a salinized landfill. Sci. Total Environ. 572, 1132–1137.
Morcillo, M., Chico, B., Mariaca, L., Otero, E., 2000. Salinity in marine atmospheric corrosion: its dependence on the wind regime existing in the site. Corros. Sci. 42, 91–104.
Muchate, N.S., Nikalje, G.C., Rajurkar, N.S., Suprasanna, P., Nikam, T.D., 2016. Physiological responses of the halophyte *Sesuvium portulacastrum* to salt stress and their relevance for saline soil bio-reclamation. Flora 224, 96–105.
Rozema, E.R., Gordon, R.J., Zheng, Y., 2016. Harvesting plants in constructed wetlands to increase biomass production and Na^+ and Cl^- removal from recycled greenhouse nutrient solution. Water Air Soil Pollut. 227, 136.
Sargeant, M.R., Tang, C., Sale, P.W.G., 2008. The ability of Distichlis spicata to grow sustainably within a saline discharge zone while improving the soil chemical and physical properties. Soil Res. 46 (1), 37–44.
USDA, 2018. Saltgrass *Distichlis spicata* (L.) Greene plant fact sheet. Retrieved from. https://plants.usda.gov/java/.
Wakeel, A., 2013. Potassium-sodium interactions in soil and plant under saline-sodic conditions. J. Soil Sci. Plant Nutr. 176, 344–354.
Yensen, N., Biel, K., 2006. Soil remediation via salt-conduction and the hypotheses of halosynthesis and photoprotection. In: Khan, M.A., Weber, D. (Eds.), Ecophysiology of High Salinity Tolerant Plants. Springer, Dordrecht, Netherlands, pp. 313–344.
Yuan, F., Leng, B., Wang, B., 2016. Progress in studying salt secretion from the salt glands in recretohalophytes: how do plants secrete salt? Front. Plant Sci. 7, 1–12.