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Spatio-temporal distribution of atmospheric chloride deposition over a small island

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Abstract

Estimating chloride deposition rates is important for several applications. The conservative nature of chloride makes it a widely used natural tracer for hydrological investigations, such as groundwater recharge estimations and catchment salt balances. Additionally, chloride deposition is used to identify the sources and pathways of air pollutants as well as predict the likelihood of corrosion in infrastructures. The variability of atmospheric chloride (Cl^-) deposition is a crucial consideration in hydrological investigations, particularly in coastal areas where it can fluctuate greatly. On small islands, spatial variations of chloride deposition have been found to be particularly strong, which may lead to large estimation errors when assuming point measurements are representative of larger areas. This paper aims to improve knowledge of the factors affecting chloride deposition's spatial and temporal variability in small islands using Norfolk Island (South Pacific) as a case study. A monitoring network consisting of 15 open-site rainwater collectors was installed, from which 275 rainwater samples were collected for the period September 2020–July 2022. Total deposition rates for this period varied from 22.7 to 36.7 mg m^{-2} d^{-1} for sites located more than 400 meters from the coast. A higher variation is observed closer to the coast, with values varying from 50.9 to 1022.7 mg m^{-2} d^{-1} for sites located between 50 and 200 m from the coast. Statistical analysis shows that distance from the nearest point on the coast and elevation above sea level are significant factors contributing to spatial variability of chloride deposition, whereas period-total rainfall depth and period-average maximum daily wind gust speed significantly affect the temporal variability. This study contributes to knowledge on chloride deposition variability as well as the understanding of factors contributing to this variability in small islands. Additionally, a double exponential model for spatial distribution of chloride deposition in small islands is proposed.

KEYWORDS
atmospheric deposition, chloride mass balance, coastal areas, Norfolk Island, Pacific islands, rainfall, sea spray, Spatio-temporal modelling

1 | INTRODUCTION

Chloride (Cl^-) is a chemical element that occurs naturally in the atmosphere and is mostly derived from the ocean (Eriksson, 1959; Gustafsson & Hallgren Larsson, 2000). In coastal areas, the ocean is the main source of chloride deposition, while in inland continental areas, other sources, such as salt lakes (Short et al., 2017), gaseous chloride (Keywood et al., 1997) and human activities (i.e., road salts, biomass burning, coal combustion and waste incineration) (Yang et al., 2018) can contribute to chloride deposition. Chloride is deposited on the land surface in two ways: dry and wet deposition. Dry deposition occurs when salt particles and gases are carried by wind and settle on the land surface (flat terrains, hills, vegetation, buildings, etc.), while wet deposition occurs when chloride in the atmosphere is incorporated in precipitation (e.g., rain, hail, snow, fog or cloud) (Boyce, 1954; Cole & Paterson, 2004; Feliu et al., 1999; Gong et al., 1997; Gustafsson & Franzén, 1996). Bulk deposition refers to the total amount of deposition and can be measured directly using continually open rainwater collectors that capture both precipitation and particles deposited from the atmosphere. These collectors are typically installed in open areas or under vegetation (throughfall bulk deposition) (Feth, 1981).

Understanding chloride deposition holds significant importance for a range of practical applications. Given its stability, conservative nature, and high solubility, chloride serves as a valuable tracer in hydrological investigations, facilitating the comprehension of water sources, movement and solute transport in the environment (Alcalá & Custodio, 2014; Crosbie et al., 2018; Feth, 1981; Kirchner et al., 2010; Nava et al., 2020; Neal & Kirchner, 2000). Although long-term chloride data sets in rivers and groundwater are commonly available to support catchment-scale tracer studies, long-term deposition data sets are rare, especially those representing spatial variability over catchments. Some applications of chloride deposition data in hydrological studies include groundwater recharge estimation using chloride mass balance (Anderson, 1945; Crosbie et al., 2018; Eriksson & Khunakasem, 1969; Ordens et al., 2012), the establishment of catchment salt balances (Anderson et al., 2019; Cartwright et al., 2013a), evaluation of catchment disturbances to hydraulic equilibrium (Cartwright et al., 2013b) and gaining insights into storage and mixing processes across catchments (Kirchner et al., 2010). Additionally, chloride bulk deposition measurements have been used in studies on metallic corrosion predictions (Chen et al., 2021; Wallinder et al., 2014); interactions of marine aerosols with concrete structures (Meira et al., 2010); understanding the capacity of canopies to capture aerosols (Beier & Gundersen, 1989; Ulrich, 1983) and as a tracer on understanding the origin and transport of atmospheric pollutants (Kang et al., 2003).

Chloride deposition rates can vary significantly in space and time. Previous studies identified several controlling factors, including: (i) distance from the coast (Barr, 1914; Biggs, 2004; Davies & Crosbie, 2018; Delalieux et al., 2006; Guan et al., 2010; Gustafsson & Hallgren Larsson, 2000; Hutton, 1976; Keywood et al., 1997; Meira et al., 2003; Wilkins et al., 2022).

(ii) weather conditions, such as rainfall depth, wind speed and wind direction (Gustafsson & Hallgren Larsson, 2000; Meira et al., 2006); (iii) topography (Guan et al., 2010); (iv) vegetation cover, including vegetation type, vegetation structure and leaf area index (Bresciani et al., 2014; Deng et al., 2013; Pyatt & Haywood, 1989; Zhang et al., 2017); and (v) sea spray generation due to the action of wind and waves at the coast (Malalis, 2006). The influences of these factors are highly dependent on the characteristics of the study site.

Distance from the coast is an important factor contributing to chloride deposition rates in coastal areas, for both continental- and small-scale studies (Bresciani et al., 2014; Davies & Crosbie, 2018; Guan et al., 2010). As marine aerosol particles are transported inland from the ocean, larger particles tend to settle out first by gravity, while small particles can travel longer distances, which leads to higher deposition in coastal regions (Barr, 1914; Bresciani et al., 2014; Griffiths & Orians, 2004; Guan et al., 2010; Randall, 1970). Previous investigations found that chloride deposition decreases exponentially with distance from the coast and, after a certain distance inland, chloride deposition tends to have lower spatial variation (Bresciani et al., 2014; Guan et al., 2010; Gustafsson & Franzén, 1996; Hutton, 1976; Keywood et al., 1997; Meira et al., 2006).

The influence of topography and vegetation on chloride deposition variability was found to be significant in small-scale studies. In Mount Lofty (Australia), a small-scale study located in a coastal hilly area, terrain aspect and slope contributed to 8% of spatial variability (with the largest contributor being distance from the coast with 65%) (Guan et al., 2010). Vegetation enhances chloride deposition rates by intercepting particles such as aerosols, cloud droplets and fog (Eriksson, 1959; Ulrich, 1983). Many studies have shown that chloride deposition under vegetation, through throughfall and stemflow, is higher than in open sites (Bresciani et al., 2014; Deng et al., 2013). Understanding these spatial and temporal variations is important when generalizing point measurements of chloride deposition for use at larger scales such as a catchment salt balance (Jolly et al., 2001). The errors introduced in generalizing point measurements may be small in inland areas far from the coast (i.e., 100's of km) but are very significant in near coastal environments with high gradients in chloride deposition (Guan et al., 2010).

On small islands, it is expected that chloride deposition is strongly influenced by the ocean. Small islands are here defined as islands with a land area ≤2000 km^2 or maximum width ≤ 10 km (Falkland & Custodio, 1991). Several studies investigated chloride deposition in small islands (Kang et al., 2003; Malalis, 2006; Marrero Díaz, 2010; Naranjo et al., 2015; Shotyk, 1997; Suzuki & Robertson, 2011), and are summarized in Table 1. Bulk deposition values in these studies varied significantly among the islands, ranging from 0.8 to 319 mg m^{-2} d^{-1}. Herrera Lameli (2001) and Marrero Díaz (2010) found extremely high chloride deposition values when sites are very close to the coast (~50 m) in Tenerife and Fuerteventura (Canary Islands). The authors attributed these values to dry deposition due to marine aerosols. In Gran Canaria (Canary Islands), chloride deposition decreased at a rate of 0.82 mg m^{-2} d^{-1} km^{-1} from coastal to highland regions (Naranjo et al., 2015). Previous studies in Oahu (Hawaii)

TABLE 1 Previous chloride deposition estimates in small islands.

Island name	Island size (km^2)	Rainfall ($mm.Yr^{-1}$)	Approx. Distance from the coast or distance from the coast range (m)	Elevation or elevation range (m ASL)	Deposition type	Number of monitoring open sites	Number of sampling rounds	Total period (months)	Chloride concentration ($mg\ l^{-1}$)	Chloride deposition ($mg\ m^{-2}\ d^{-1}$)	Reference
Norfolk Island (location of this case study)	35.7	1282	2000	128	Wet deposition	1	10	–	0.5–11.4	–	Abell and Taylor (1981)
Norfolk Island (location of this case study)	35.7	1282	404–2063	–	Bulk deposition (rainwater tanks)	6	1	–	8.2–20.0	20.5–51.2	Petheram et al. (2020)
Betancuria area, Fuerteventura Island, Canary Islands, Spain	1660	114	50–15 000	2–680	Bulk deposition	6	6	–13	3.2–265.9	8.1–319.2	Herrera Lameli (2001)
Las Cañadas Caldera, Tenerife, Canary Islands, Spain	216	560	165–18 727	60–2149	Bulk deposition	22	18	17	2.2–27.3	9.0–122.5	Marrero Díaz (2010)
North-eastern Gran Canaria	312	387	–	315–1370	bulk deposition	10	18–54	72	6.7–28.9	11.5–25.2	Naranjo et al. (2015)
Oahu (Hawaii, USA)	1545	–	–	-	dry deposition	5-6	10-31	6-30	–	6.8–2883	Malalis (2006)
Tuitila, American Samoa	142.3	3200	50	76	Wet deposition	1	124	–	0.1–1	–	Dayan and Nelson (1988)
Jeju City, Jeju Island, South Korea	1846	2153	9000–15 000	473–1100	Wet deposition	2	161–281	62	1.0–2.3	7.7–9.1	Kang et al. (2003)
Lopez Island, Washington, USA	76.4	657	1000–2000	–	Bulk, wet and dry deposition	3	21	27	0.3–27.4	2.4–4.0	Orr et al. (2002)
Olkiluoto Island, Finland	9	542	1000	–	Bulk deposition	1	–	12	–	0.8–1.6	Haapanen (2009)
Okinawa Island, Japan	1207	1769	2000	–	Wet deposition	1	189	23	12.4	–	Sakihama et al. (2008)
Island of Foula (Islands, Scotland	15	1000–1500	–	–	–	1	15	22	6.2–305.5	–	Shotyk (1997)

suggest that wind direction, wind speed and wave size possibly contribute to differences in dry deposition rates between coastal sites (Malalis, 2006; Suzuki & Robertson, 2011). The same variables also cause deposition rates to vary over time. For instance, Sakihama et al. (2008) noted higher deposition during cyclones, typhoons and wet seasons. Waterloo et al. (1997) had similar findings for the Viti Levu Island (Fiji), although with an area >10 000 km^2 it falls outside of our definition of small islands. Most studies that estimated chloride deposition in small islands have focused on the origin and chemical characterization of precipitation, and a few have focused on recharge estimation, corrosion predictions and mass balance of atmospheric inputs.

Therefore, accurate chloride deposition data have a pivotal role in advancing hydrological understanding, especially in small islands with limited monitoring data. Despite earlier attempts, an incomplete understanding persists regarding the spatio-temporal distribution of chloride deposition in small islands. This paper addresses this gap by conducting an in-depth examination of the spatio-temporal variability and the relative importance of drivers influencing this variability. Furthermore, it tackles knowledge deficiencies related to the limited resolution of monitoring networks in previous small island studies, specifically concerning the number of sites and sampling periods.

The primary objective of this study is to enhance knowledge on the variability of chloride bulk deposition in small islands by comprehensively examining the influence of spatial and temporal factors through statistical and regression analysis. The study analyses data from a relatively dense network of monitoring sites and a substantial number of sampling periods over approximately 2 years (15 sites covering 35.7 km^2 across 25 sampling periods). It introduces a new spatio-temporal model for long-term deposition that guides applications across a wide range of small islands. Furthermore, the research compares chloride deposition measurements from this study to those previously reported for small islands. This study uses Norfolk Island as a case study, a very small island (35.7 km^2) part of the external territory of Australia in the South Pacific Ocean. This paper specifically focuses on chloride deposition measurements at open sites only (no nearby canopy or other objects). A parallel investigation is underway to explore the influence of vegetation on chloride deposition variability in Norfolk Island.

2 | STUDY AREA

Norfolk Island is a small volcanic island (35.7 km^2) located in the South Pacific Ocean between New Zealand and Australia (Figure 1). The island has a wide plateau approximately 100 m above sea level (ASL) that rings the peaks of Mount Pitt and Mount Bates, which have elevations of 316 and 318 mASL respectively. The coastline mainly consists of cliffs up to 100 mASL high along the northwest and up to 50 mASL high in the southeast. There is 1.5 km long, 0.5 km wide lowland on the southern side of the island at approximately 20 m ASL (Abell & Falkland, 1991). The island is largely comprised of basaltic rocks and tuffs that are weathered deeply and highly permeable soils (Petheram et al., 2020). The native vegetation of Norfolk Island is categorized as subtropical rainforest, primarily comprising of Norfolk pines (*Araucaria heterophylla*), hardwoods, palms and tree ferns (Council & TierraMar, 2021; Mills, 2008, 2012). Additionally, a substantial portion of the island is characterized by the prevalence of invasive shrub species, with African olive (*Olea europaea*), cherry guava (*Psidium cattleianum*), and Hawaiian holly (*Schinus tenebinthifolius*) being the predominant invasive species (Petheram et al., 2020).

Norfolk Island's climate is subtropical and temperate, with mean monthly maximum temperatures ranging from 18.4 to 25.0 °C and mean monthly minimum temperatures ranging from 13.3 to 19.8 °C (based on measurements from 1939 to 2022). The mean annual rainfall is 1288 mm (based on measurements from 1890 to 2023), with most rainfall occurring from May to August (Bureau of Meteorology, 2023). The annual rainfall fluctuates by more than 500 mm due to climate influences, such as El Niño–Southern Oscillation (ENSO) (Petheram et al., 2020). Dirks et al. (1998) found considerable spatial variation in accumulated rainfall with inter-site differences in the 1990–1993 average rainfall of up to 30%. These variations were attributed to wind patterns, with higher accumulations tending to occur leeward of Mount Pitt and Mount Bates and lower accumulations windward (Dirks et al., 1998). Mean annual evaporation for the period 2010–2019 was estimated to be 883 mm (Petheram et al., 2020). The predominant wind directions are east and southeast (1932–2022), with a long-term mean 9 am wind speed of 15.7 km h^{-1} and 3 pm wind speed of 18.2 km h^{-1} (1939–2010) (Bureau of Meteorology, 2023).

Two studies have previously measured rainwater chloride concentrations in Norfolk Island. From 1977 to 1980, nine rainwater samples were collected from one rainfall collector located about 2 km away from the coast (Abell & Taylor, 1981). Each sample represented a 24-h period in which a major rainfall event occurred (which may include some hours before and after rain). The focus on rainfall events indicates that the dry deposition component of these samples was minimal. Chloride concentration varied between 0.5 and 11.4 mg L^{-1}, with the lower concentrations observed during periods of heavier rainfall (Abell & Taylor, 1981). Petheram et al. (2020) estimated average chloride deposition by analysing samples from six rainwater tanks, assuming that the chloride concentrations represent the previous 12 months of rainfall. They found an exponential relationship between distance from the coast and chloride deposition. The main limitations and uncertainties associated with these studies are the short periods of data collection, a limited number of sampling points and a lack of investigation of the factors influencing deposition. Maurin et al. (2021) estimated chloride deposition by applying the equation developed by Hutton (1976) that describes the relationship between distance from the coast and chloride deposition for a coastal area in Southeast Australia. Maurin et al. (2021) highlight that a better estimation method is needed for the island context.

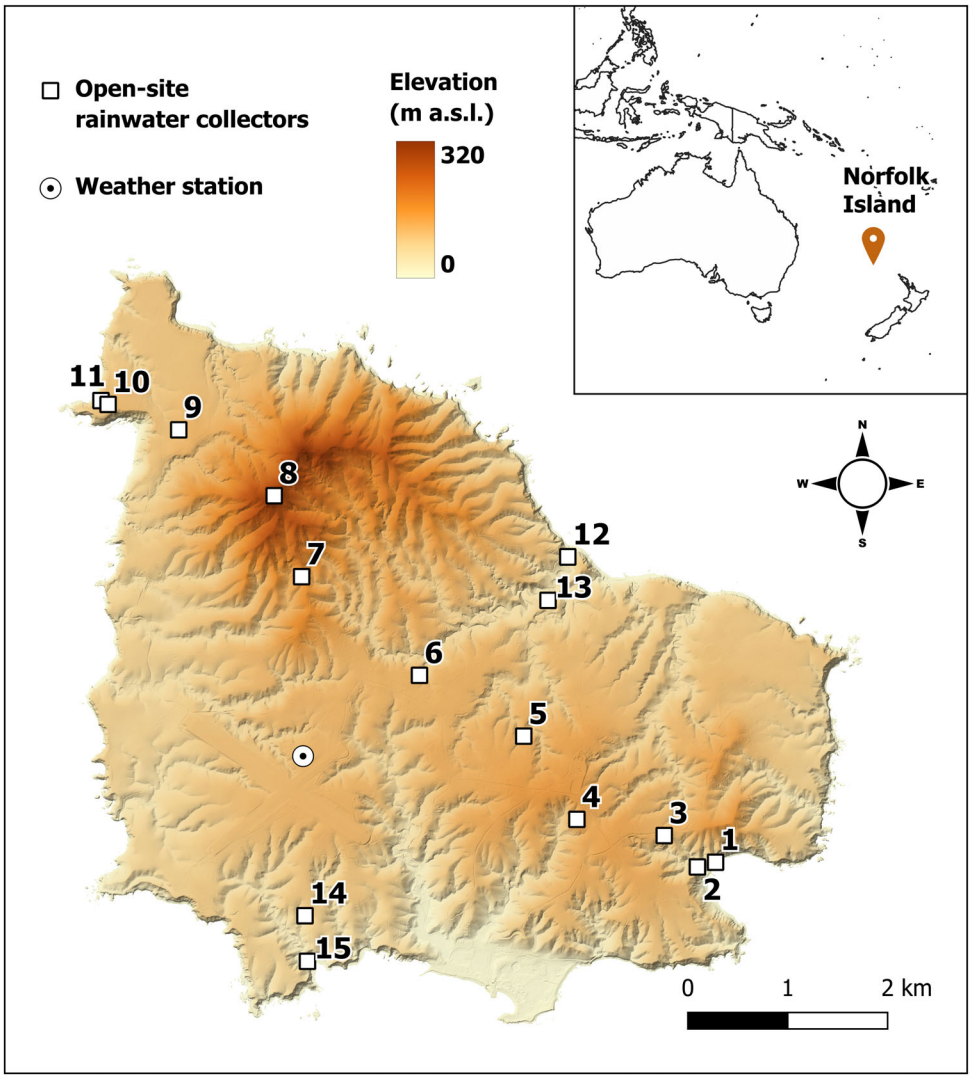

FIGURE 1 Location plan, island topography, and locations of rainwater collectors and BOM weather station.

3 | METHODOLOGY

3.1 | Monitoring network

The collection of chloride deposition and rainfall data involved the installation of rainwater collectors designed to remain continuously open to the atmosphere, allowing the capture of bulk deposition (a combination of both dry and wet deposition). The monitoring network comprised 15 open-site rainwater collectors, with 11 strategically distributed along an approximately straight line corresponding to one of the long-term prevailing wind directions, from southeast (SE) to northwest (NW) (Figure 1). These collectors were installed in September 2020 (dates are in Table A1). In October 2021, a few collectors were removed (sites 1, 4, and 7 in Figure 1) and new collectors were installed (sites 12, 13, 14, and 15) in the southwest-northeast direction. This change was useful to compare chloride deposition along the two orthogonal transects. The main criteria for the selection of sites were distance from the coast, prevailing wind direction, access practicality and safety, and landowner permission. At 10 sites, the collectors were installed no nearer to a surrounding object (e.g., trees, buildings) than the height of that object (GNIP, 2014). For the other five sites, this criterion could not be met while also meeting access criteria, due to the presence of dispersed vegetation, houses and hilly areas, which are characteristic of the island. Further details such as height of nearby objects are found in Table A2.

Rainwater collectors consisted of a funnel draining to containers (volume 2.5–5 L) held 1 m above the ground in a vertical PVC pipe set into the ground (Figure 2). Sites 12 and 15 have slightly different collector types since they were re-utilized from a previous project (Figure A1). Plastic mesh was placed on the funnel to stop insects, leaves and other debris from falling into the collector. The collectors were filled with a thin (~1 cm thick) layer of liquid paraffin oil to avoid water evaporation (IAEA, 2014).

3.2 | Sample collection and laboratory analysis

The rainwater samples were collected continuously from September 2020 to July 2022. Rainwater was allowed to accumulate in collectors over periods ranging from days to weeks before sample collection and

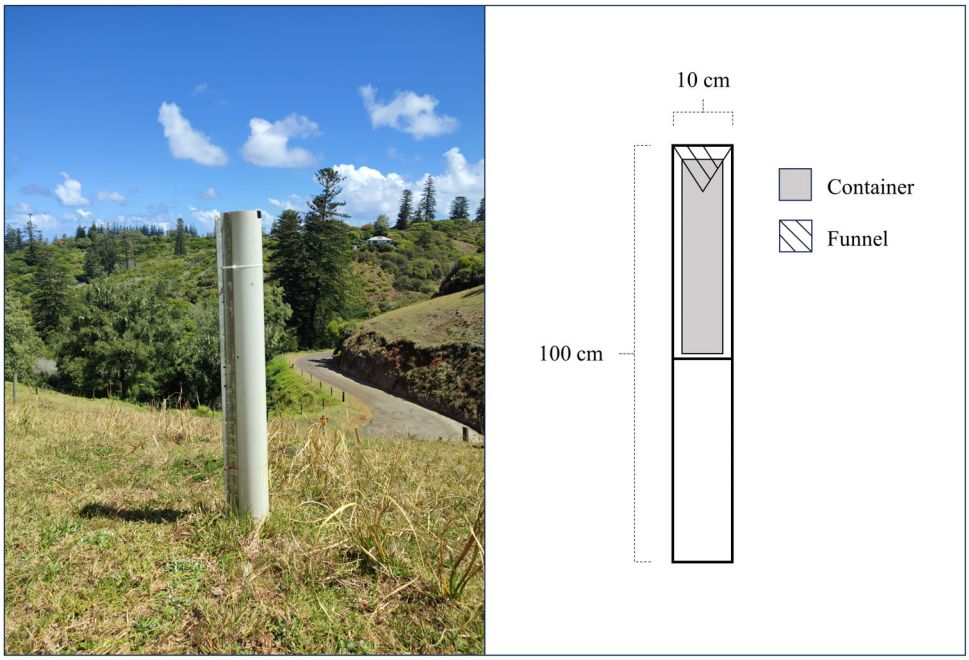

FIGURE 2 Rainwater collector at site 3 and schematic diagram of rainwater collectors.

resetting. The duration varied based on rainfall depths, weather conditions, and logistical considerations (refer to Table A1 for comprehensive period details). The objective was to capture diverse weather conditions while concurrently ensuring a minimum of 50 mL of rainwater for sampling and preventing sample overflow. The sampling period was consistent within 24 h for each site. At the conclusion of each sampling period, the rainwater volume in each collector was measured, and a minimum of 50 mL was transferred to clean 100 mL containers. Resetting the collectors involved emptying, rinsing with distilled water, and reinstating the paraffin oil layer.

Considering the long distance from a laboratory facility in the mainland and the quarantine requirements for sample transportation to mainland Australia, the samples were analysed at the local Council laboratory on Norfolk Island. Sample preparation involved separation of the paraffin oil from the water sample and filtering using 11 μm paper filters. Chloride concentration for each sample was measured by direct potentiometry using a Chloride Ion-selective Electrode (ISE) connected to smartCHEM-ion3 Ion/pH/mV/Temp bench meter (TPS, Australia). To ensure accuracy, an Ionic strength adjustment solution was added to the samples and standard solutions. The ISE probe calibration was checked before each day of laboratory analysis and calibrated as needed (∼bimonthly) using standard chloride solutions.

3.3 | Data analysis

The primary variable analysed was the chloride deposition rate (D_{BP}) (mg m^{-2} d^{-1}), which was calculated using chloride concentration (C_{BP}) (mg l^{-1}), sample volume (V) (l), funnel area (A) (m^2) and period length (t) (days):

$$D_{BP} = \frac{C_{BP}.V}{A.t} \quad (1)$$

The variation of D_{BP} over time and space, and its relationships with distance from the coast, elevation, rainfall, wind speed and wind direction were assessed. First, the spatial factors were assessed by plotting total rainfall depth and total deposition against site distance from the nearest point on the coast and elevation above sea level. The effect of prevailing wind direction on spatial variation of chloride deposition was investigated by plotting the estimated distance from the coast facing the east wind (prevailing wind direction during data collection period) and chloride deposition. Following spatial analysis, chloride deposition in sites was plotted against period-total rainfall depth, period-average daily wind speed and period-average maximum daily wind gust speed to determine temporal effects. Wind gust speed was included to test the hypothesis that deposition is affected by sea spray in extreme wind conditions that are not well represented by average wind speeds. The wind data were obtained from the BOM weather station (location in Figure 1), while all other data are site-specific. Since rainfall and chloride deposition data are right-skewed (Figure A2), they were log-transformed.

Considering the high spatial variability of chloride deposition and the site dependence of temporal effects, it is useful to analyse in further detail the spatio-temporal effects. This is approached in two ways: (1) using multiple regressions to define the relation between chloride deposition and temporal factors at each site and comparing regression results between sites; and, (2) fitting a spatio-temporal multiple regression model that includes both spatial and temporal factors. To infer the relative importance of each variable in the regression analysis, the data were standardized to have zero mean and standard deviation of one.

4 | RESULTS

In total, 275 rainwater samples were collected. A few collectors were damaged by wind, vandalism and animals during the data collection period, meaning that approximately 2% of the samples were omitted from the analysis. The total site-average rainfall over all sampling periods (9 September 2020 to 17 July 2022), calculated by summing site-average rainfall over sampling periods, was 2298 mm (equivalent to 1286 mm year^{-1}), compared to 2557 mm measured at the BOM weather station over the same period. This difference is consistent with the spatial variations seen in Table A2. The rainfall in the first year of sampling was 57% lower than in the second year. Supplementary information summarized by each site and by each period is found in Tables A1 and A2.

4.1 | Spatial analysis

To understand the influence of spatial factors on chloride deposition, the results are shown in Figure 3 separately for each sampling year. As sites 12–15 were present only in year 2, and sites 1, 4, and 7 only in year 1, combining the 2 years in Figure 3 would lead to confounding temporal effects (temporal effects are addressed in Section 4.2).

Figure 3 shows results over the sampling sites for chloride deposition and rainfall. Further details including number of samples for each site, distance from the coast, elevation and chloride concentration are found in Table A2. Accumulated rainfall among sites varied from −24% to +17% from the site-mean and is generally slightly higher for sites located nearer the middle of the island at higher elevations (sites 4, 5, 6, and 7) with the notable exception of sites 12, 14, and 15 in

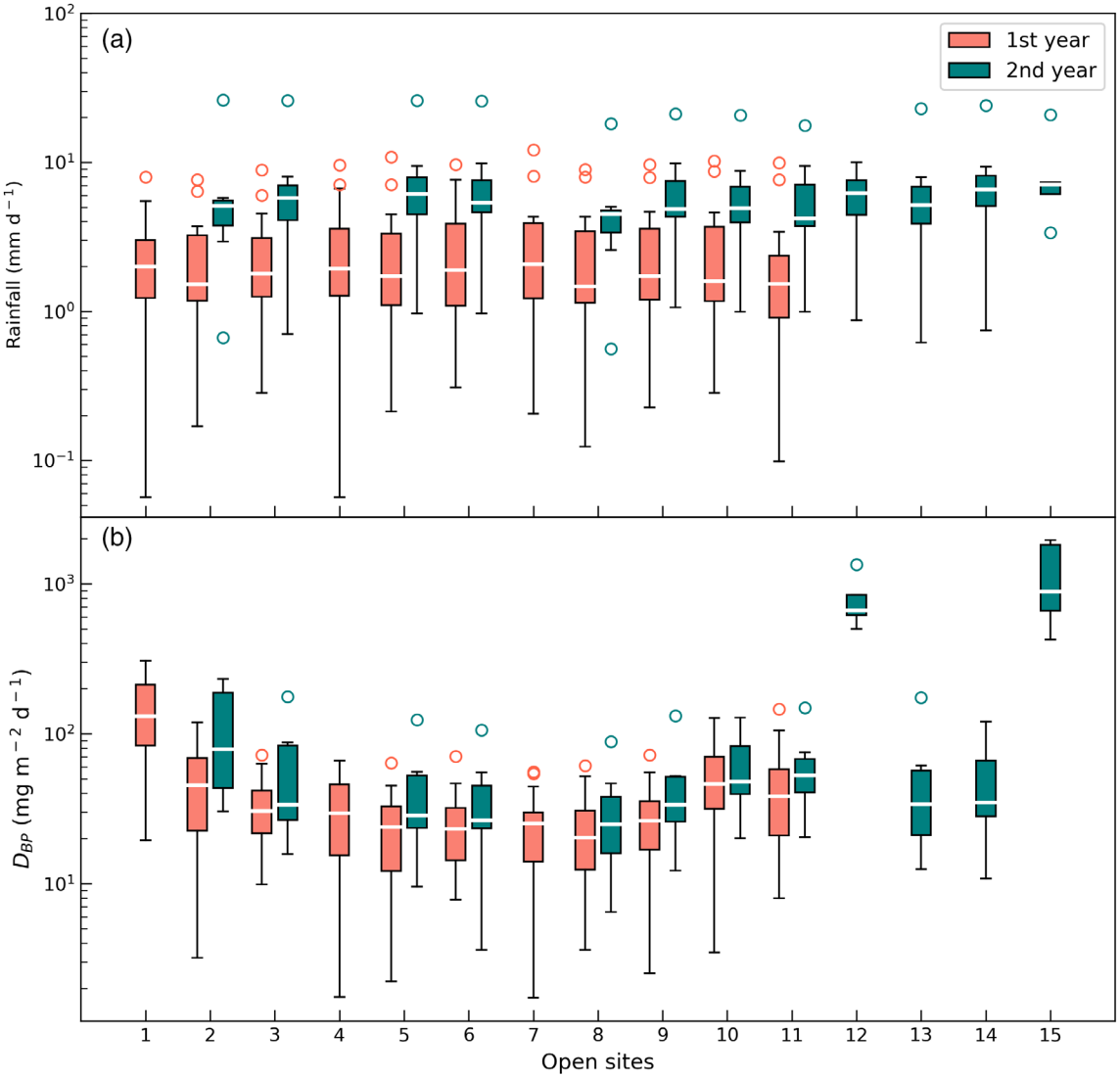

FIGURE 3 Spatial variation of rainfall and chloride deposition by site. Rainfall is the period-total rainfall (mm d^{-1}) and D_{BP} is the period-total chloride bulk deposition (mg m^{-2} d^{-1}). The data is shown separately for the first year (9 September 2020–1 October 2021) and the second year (2 October 2021–17 July 2022) of sampling. The box and whisker plots show the variation over the sampling periods for each site, showing 5, 25, 50, 75, and 95 percentiles and any outlying values. Numbering in the x-axis corresponds to sites depicted in Figure 1.

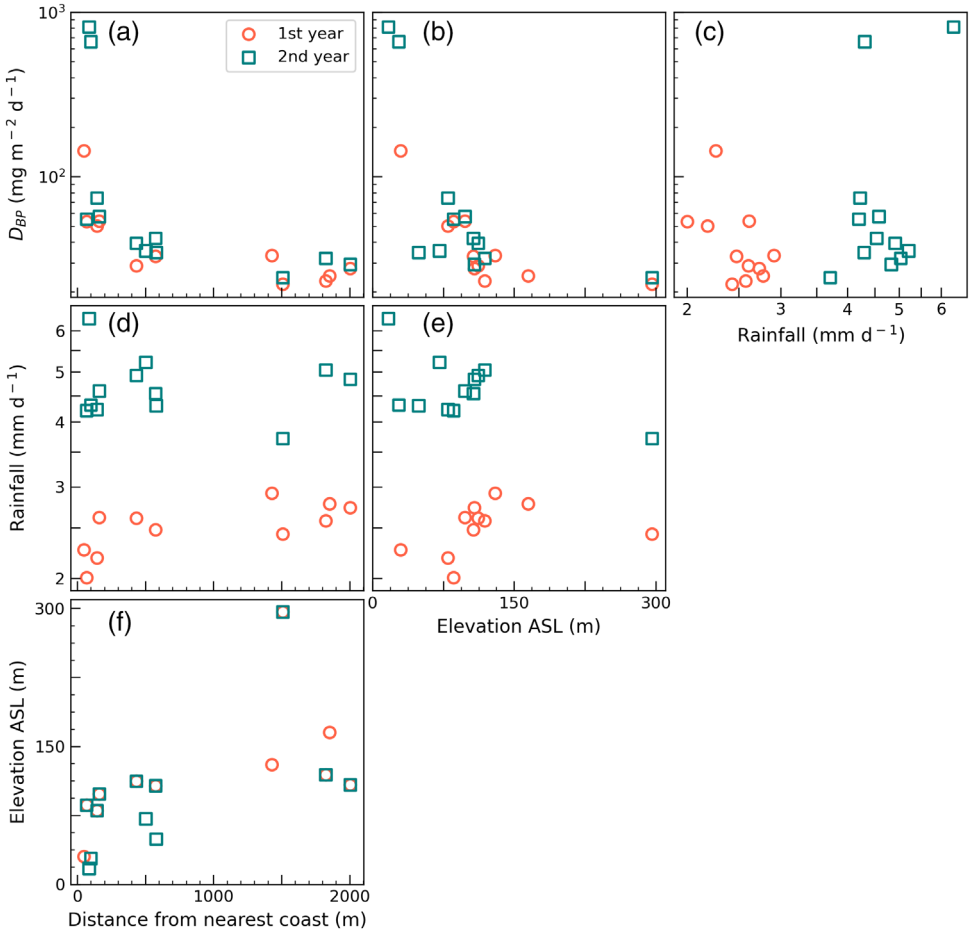

FIGURE 4 Scatter plots of spatial variables. D_{BP} (chloride deposition in mg m^{-2} d^{-1}) and Rainfall (in mm d^{-1}) represent the total deposition and accumulated rainfall at each site, respectively. The results are presented separately for each sampling year. Of a total of 283 samples, 8 samples were missed from sites 2, 7, 12, and 15 combined. The data for all sites as well as the missing samples are detailed in Table A2.

year 2, which had high median rainfall despite being near to the coast. This may be partly because sites 12 and 15 captured a smaller number of year 2 periods (4 and 5, respectively; Table A2) due to issues to access sites and damaged collectors; furthermore, year 2 presented a higher volume and higher variation of rainfall among sites compared to year 1, due to the higher occurrence of heavy rainfall events. Site 8 is located inland near the top of Mount Pitt (318 m ASL) but presented low rainfall. Since Site 8 is exposed to the wind from all directions, this may have led to an under catch of rain by the rainwater collector.

While rainfall is relatively uniform over the island and tends to be slightly higher for sites located nearer the middle of the island, chloride deposition varies more strongly and is higher at sites close to the coast (sites 1, 2, 10, 11, 12, and 15). For example, average chloride deposition was 26 mg m^{-2} d^{-1} for sites located approximately between 1500 and 2000 m from the coast, 35 mg m^{-2} d^{-1} for sites located between 400 and 600 m from the coast, and 407 mg m^{-2} d^{-1} for sites located between 50 and 200 m from the coast.

Figure 4 shows all relations between chloride depositions (mg m^{-2} d^{-1}), distance from the nearest coast (*m*), elevation (*m*), and rainfall (mm d^{-1}). Here, for each site, chloride deposition and rainfall represent the total sum over sampling periods divided by the number of sampling days. Rainfall presents a generally positive relationship (Figure 4d,e) with distance from the coast and elevation with

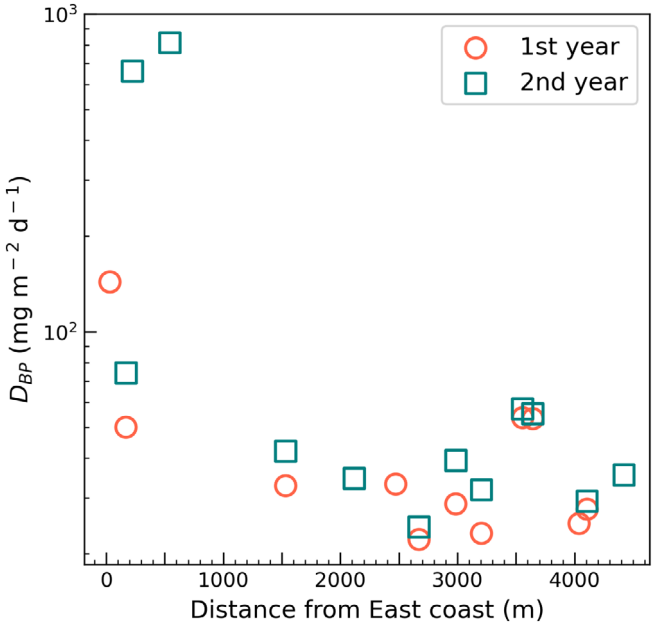

FIGURE 5 Relationship between distance from the east (prevailing wind) coast and chloride deposition.

exceptions of sites 8, 12, 14, and 15 as previously discussed. The spatial differences in rainfall (mm d^{-1}) do not seem to consistently affect the spatial variation of chloride deposition (Figure 4c). This implies

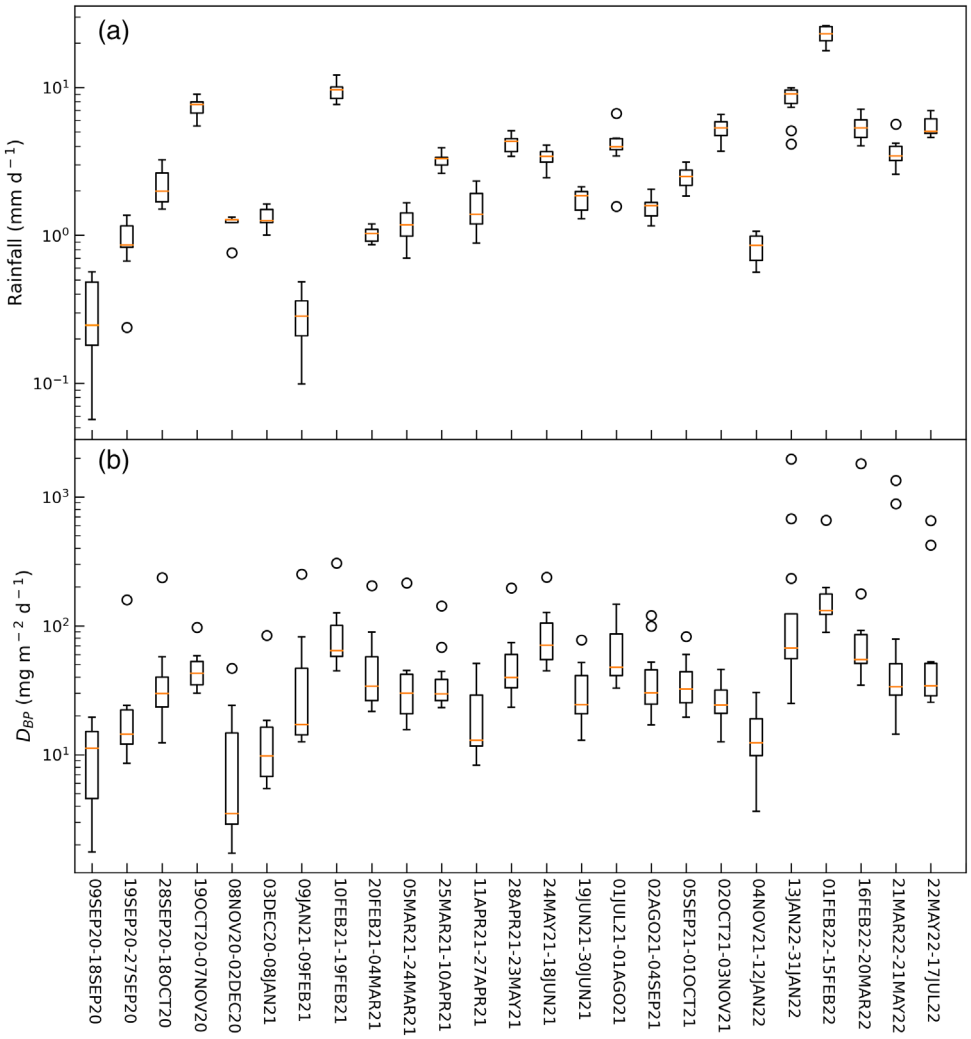

FIGURE 6 Rainfall and chloride deposition results over sampling periods. D_{BP} is chloride deposition (mg m^{-2} d^{-1}).

that the strong effect of distance from the coast on chloride deposition is offsetting any spatial effect of rainfall. Elevation is also strongly related to chloride deposition (Figure 4b), but this may be influenced by the positive relationship between elevation and distance from the coast (Figure 4f).

The influence of prevailing wind direction in the spatial variation of chloride deposition is investigated in Figure 5. Since the east was the prevailing wind direction of the data collection period (Figure A3), it is expected that the east coast receives more marine aerosols when compared to other coasts. The results in Figure 5 confirm that distance from the east coast greatly influences the chloride deposition.

4.2 | Temporal analysis

The rainfall and deposition data were averaged over sites for each sampling period. Results are plotted in Figure 6 and listed in Table A1. Sites 1, 12, and 15 were excluded from the site-averaged results in Table A1 because, due to their high deposition values, they would dominate these values and hence dominate the interpretation of temporal effects. These sites are very close to the coast (50, 100, and 90 m, respectively) and are assumed to be strongly influenced by local effects of near-ocean sea spray. In Figure 6, these sites are shown as outliers.

Figure 6 shows that the rainfall and chloride deposition vary strongly over time and that periods with higher inter-site variation in chloride deposition are not necessarily the periods with higher inter-site variation in rainfall. These features indicate the presence of additional temporal and spatio-temporal factors.

Figure 7 presents scatter plots of chloride deposition against rainfall, average wind speed and average maximum wind gust speed. The plots indicate that all three variables may significantly influence the temporal variability of chloride deposition and their influence can vary depending on each site location. Additional scatter plots presenting all relations between temporal variables and chloride deposition by each site individually are found in the appendix (Figures A5–A9).

4.3 | Spatio-temporal analysis

The spatio-temporal analysis involved two steps: (i) a temporal regression analysis at each site separately and a comparison of regression

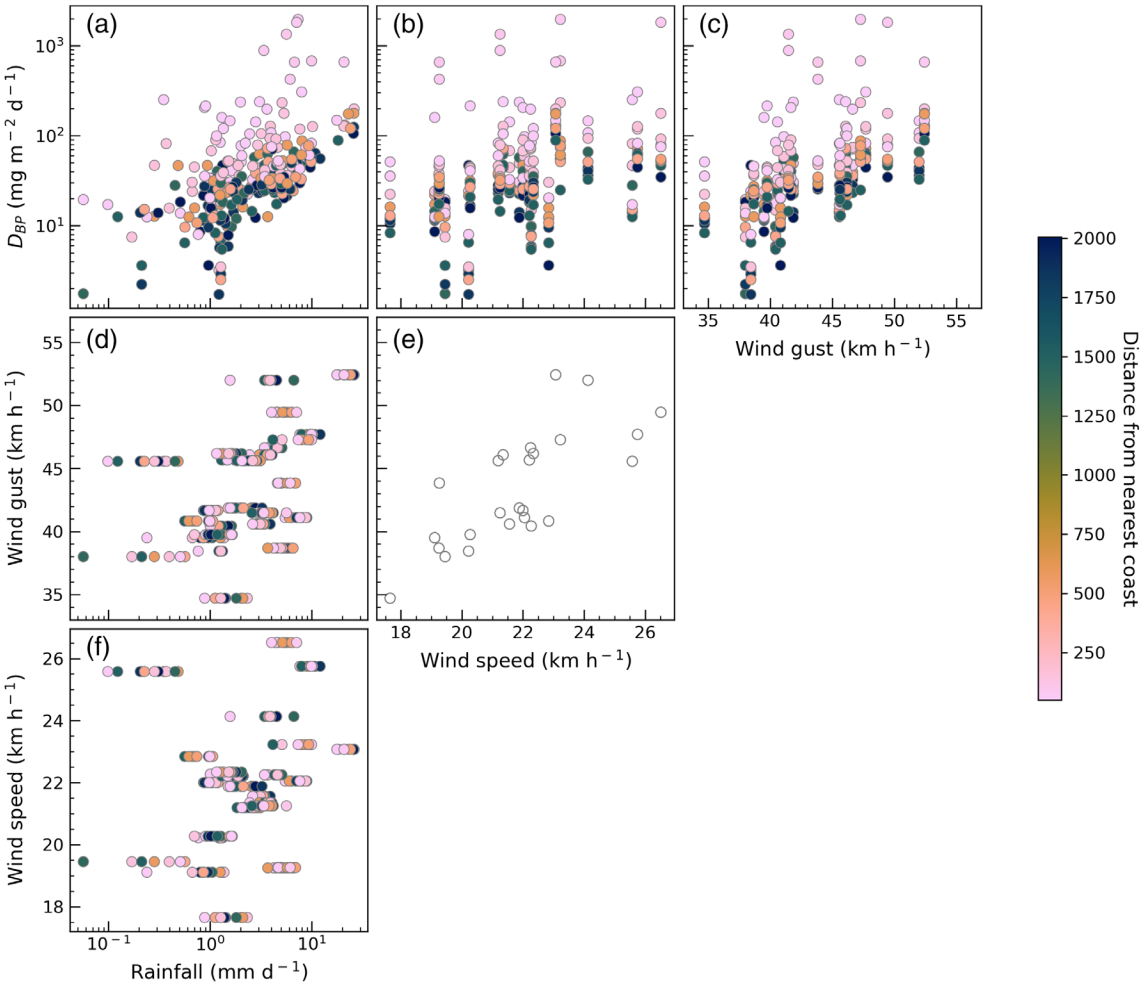

FIGURE 7 Scatter plots illustrating the correlation between temporal variables and chloride deposition, along with a depiction of how the proximity to the coast influences these relationships. D_{BP} (chloride deposition in mg m^{-2} d^{-1}) and Rainfall (mm d^{-1}) represent the results at each sampling period for all sites separately, while Wind speed (period-average daily wind speed in km h^{-1}) and Wind gust (period-average daily maximum wind gust in km h^{-1}) represents results for each period attributed for all sites.

results between sites; (ii) a spatio-temporal multiple regression model that includes both spatial and temporal factors. Please see further details in section 3.3.

The multiple regression results applied per site show that both rainfall and average maximum wind gust speed were statistically significant for all sites (p-value <0.1), except for sites 1, 12, and 15 (Table 2). The p-values indicate that the wind gust speed has a more definite effect at sites located closer to the coast (1, 2, 3, 9, 10, 11, 13, 15), while the rainfall effect is more definite at sites located in the middle of the island (4, 5, 6, 7, 8), except for sites 12 and 14. Including average wind speed or rainfall intensity (total rainfall divided by the number of rainy days at each period) in these regression models did not significantly improve the adjusted R^2 values.

The spatio-temporal, standardized multiple regression applied to 275 samples (all available samples covering all periods and all sites) returned the following equation:

$$log_{10}D_{BP} = 0.34log_{10}R + 0.34Wg - 0.29X - 0.23E \quad (2)$$

where:D_{BP} is chloride deposition calculated at each sample [mg m^{-2} d^{-1}]R is rainfall measured at each sample [mm d^{-1}]Wg is the calculated average of daily maximum wind gust for each sampling period [km h^{-1}]X is the distance at each site from the nearest coast [m]E is the elevation above sea level at each site [m]

The p values for log10 R, Wg, E, and X show a high level of significance (p <0.00001). The adjusted R^2 value is 0.57. Removing any of the variables in Equation (2) substantially decreases this R^2 value. Replacing average maximum wind gust (Wg) with period-average wind speed reduced R^2 from 0.57 to 0.54, and including both did not improve upon 0.57. Including distance from the coast in the prevailing (east) wind direction as an additional input increases R^2 from 0.57 to 0.62.

The regression results and residual plots (Figure A4) show that sites 8 and 12, as well as period 5, can be considered outliers. Excluding these results would increase R^2 from 0.57 to 0.66. The reasons for these outliers are discussed in section 5.3.

TABLE 2 Temporal regression analysis at each site.

Site	Number of samples	Adjusted R^2	Standardized coefficient Wg	Standardized coefficient $log_{10}R$	*p*-value Wg	*p*-value $log_{10}R$
1	18	0.21	0.35	0.30	0.16	0.23
2	23	0.58	0.60	0.28	0.00	0.10
3	25	0.65	0.36	0.26	0.00	0.01
4	19	0.63	0.41	0.57	0.02	0.00
5	25	0.75	0.45	0.55	0.00	0.00
6	25	0.51	0.34	0.50	0.06	0.01
7	17	0.42	0.39	0.46	0.07	0.04
8	25	0.7	0.39	0.59	0.01	0.00
9	25	0.66	0.49	0.45	0.00	0.00
10	25	0.5	0.52	0.32	0.01	0.07
11	25	0.68	1.06	0.79	0.00	0.00
12	4	0.4	−1.60	1.89	0.34	0.30
13	7	0.91	0.66	0.38	0.02	0.09
14	7	0.93	0.46	0.61	0.03	0.01
15	5	0.48	2.08	−2.02	0.14	0.15

5 | DISCUSSION

5.1 | Spatial analysis

The rainfall spatial variability was small compared to the variations over sampling periods. Nevertheless, rainfall was generally higher in the middle of the island than closer to the coast. Site 8, located in the middle of the island, is therefore considered an outlier since it presented consistently lower rainfall amounts when compared to nearby sites. This finding is consistent with a previous investigation on Norfolk Island that found the same at a location roughly 500 m (horizontal distance) from site 8 (Stow & Dirks, 1998). Previous studies found that rainfall under catch by rainwater collectors can be significant in areas more exposed to the wind (Folland, 1988; Pollock et al., 2018). Since site 8 sits exposed near the top of Mount Pitt, this seems a plausible explanation.

Chloride deposition presented a high spatial variability, varying from 22.7 to 1022 mg m^{-2} d^{-1} across sites. This large variation agrees with those of previous studies in the Canary Islands (Herrera Lameli, 2001; Marrero Díaz, 2010; Naranjo et al., 2015). The range of 22.7–32.5 mg m^{-2} d^{-1} for five sites (sites 4, 5, 6, 7, and 8) located between 1430 and 2000 m from the coast is similar to the results of Marrero Díaz (2010), who found that chloride deposition was 29.6 and 30.4 mg m^{-2} d^{-1} for sites located at 1222 and 2034 m from the coast in the Canary Islands. In addition, a recent investigation in Norfolk Island found similar chloride deposition estimates (25.5–32.0 mg m^{-2} d^{-1}), using samples from rainwater tanks, located between 1260 and 2063 m from the coast (Petheram et al., 2020). Particularly high deposition rates (143–1022.7 mg m^{-2} d^{-1}), found at sites located northern coast (within 100 m and below 30 m ASL), are consistent with studies elsewhere (Herrera Lameli, 2001; Marrero Díaz, 2010).

The spatial analysis demonstrates that distance from the nearest coast and elevation significantly affect the spatial variation of chloride deposition, showing a rapid decrease in chloride deposition in the first 200 m from the coast and 80 m of elevation (Figure 4). In agreement with several studies (Biggs, 2004; Bresciani et al., 2014; Guan et al., 2010; Gustafsson & Hallgren Larsson, 2000; Keywood et al., 1997; Petheram et al., 2020), scatter plots suggest a non-linear negative relationship between distance from the coast/elevation and chloride deposition rate. Considering the positive relationship on Norfolk Island between distance from the coast and elevation (Figure 4f), it is difficult to identify from the figure which of these variables is dominant concerning spatial effect. In coastal areas, it is generally accepted that the combination of lower elevations and proximity to the coast increases the exposure of marine aerosols, due to the direct influence of sea spray released by breaking waves. This, therefore, explains the influence of both variables on chloride deposition.

The negative relationship between elevation and chloride deposition found in this study differs from previous studies in continental areas, where higher deposition at higher elevations is often explained by the higher rainfall found in these areas all else being equal. Hence, in this present study, it seems that the distance from the coast and associated dry deposition effect overrides the rainfall effect on spatial variability, considering that rainfall is generally higher in the middle of the island and at higher elevations. This in turn indicates that the spatial variability of rainfall presented in this study does not appear to affect the spatial variation of chloride deposition significantly. The fact that the island is small and presents low spatial variability of period-average rainfall possibly contributes to this finding. This result implies that spatial variability of chloride deposition is mostly due to differences in concentrations of chloride in bulk precipitation among sites, rather than the differences in rainfall volume. The higher

concentrations of chloride in bulk deposition close to the coast are explained by higher rates of dry deposition and/or higher concentrations of chloride in wet deposition. Based on the literature, the dry deposition component is the most likely contributor since the dry/bulk ratio significantly increases with proximity to the coast (Alcalá & Custodio, 2008).

It is expected that the prevailing wind direction facilitates the occurrence of marine aerosols and consequently causes an increase in chloride deposition for sites located closer to the coast in the prevailing up-wind direction. The results of this study show that there is a correlation between distance from the east coast and deposition, indicating that wind direction may influence the spatial variation of chloride deposition (Figure 5), in addition to the factors previously mentioned. For instance, sites 3 and 9 are located at similar distances from the nearest coast and elevations, and present similar rainfall amounts, however, site 3 is more exposed to the east coast than site 9 (Table A2; Figure 1). The fact that chloride deposition at site 3 was consistently higher than at site 9 is an indication that the prevailing easterly wind direction transports higher amounts of aerosols to site 3. For the remaining sites, differences in distances from coast, elevation and rainfall amounts preclude any interpretation of this effect (Table A2).

5.2 | Temporal analysis

Changes in rainfall, average wind speed and maximum wind gust positively correlate to chloride deposition and significantly influence temporal variations. Similarly, a few studies have found higher chloride deposition during periods with cyclones and typhoons, which are normally related to a combination of higher wind speed and rainfall (Gustafsson et al., 2000; Sakihama et al., 2008). Based on previous studies, the increase in bulk chloride deposition due to wind speed is highly related to the increase in the dry deposition component (Corvo-Pérez et al., 2015; Meira et al., 2006). This is explained by the increase in the number and size of marine aerosols in the atmosphere, which are carried by wind and eventually deposited, on the surface.

Although temporal variations in chloride deposition and rainfall are positively correlated ($R^2 = 0.76$ for averaged results) (Table A1), chloride concentration and rainfall are negatively correlated (Table A1). The negative relationship between chloride concentration and rainfall is explained by the dilution effect of particles in the rainwater sample as the volume of rainfall increases during individual rainfall events, in agreement with previous studies (Blackburn & McLeod, 1983; Guan et al., 2010; Stout, 2019). According to Blackburn and McLeod (1983), the concentration of aerosols and salt particles, captured by the rainfall in the atmosphere, is higher in the 1 min of rainfall and is diluted as rainfall volume increases during a rainfall event. The amount of dry deposition will also influence the concentration and its relationship with volume, as shown by the higher deposition rates and concentrations found near the coast (Table A2).

5.3 | Spatio-temporal analysis

This study shows that the temporal effect depends on location, especially concerning distance from the coast and prevailing wind direction. The regression results by site (Table 2) show that both rainfall and average maximum wind gust speed explain much of the temporal variation in chloride deposition for most sites. It seems that, in coastal areas, wind gust has a higher influence on chloride deposition, while in inland areas, rainfall is the dominant factor. The lower significance of rainfall for coastal sites is consistent with the previous discussion in section 5.1, regarding the increased role of dry deposition at these sites through transportation of marine aerosols.

Interestingly, the three sites located up to 100 m from the coast and up to 30 m, ASL presented high *p*-values (Table 2) for both average maximum wind gust speed and rainfall, implying that these variables may not be significant drivers of deposition. Since these three sites are located very close to the coast, it is possible that they are being affected by local wind gust speed variations, while the wind data used in the study was from the BOM weather station in the interior of the island.

The general regression results indicate that average maximum wind gust speed, rainfall, distance from the coast and elevation explain 57% of the temporal and spatial variations of chloride deposition. Including distance from the prevailing wind direction coast increases R^2 from 0.57 to 0.62, indicating that prevailing wind direction can also influence spatial variability of chloride deposition. A constraint in further incorporating the role of wind direction is the lack of spatio-temporal wind speed data, which may provide some insight into the importance of local average wind speeds or local wind gusts. While such wind data are available (to some extent) in areas covered by Doppler weather radar, this does not apply to Norfolk Island. Considering the data limitations to include the variables mentioned above, further enhancement of the spatio-temporal model was not attempted.

The regression equation using standardized variables implies that rainfall and wind gust, each with a coefficient of 0.34 in Equation (2), are equally important controls on temporal variation of chloride deposition. Distance from the coast and elevation also have comparable coefficient values (0.29 and 0.23). While there is some correlation for each of these pairs (see Figures 4 and 7), the presence of independent effects is unambiguous, since removing any of the variables leads to substantial reductions in R^2.

Residual analysis shows a cluster of extremely negative residuals (the model overestimates the measured chloride deposition) across all sites during period 5 in November 2020 (Table A1), the reason for which is unknown.

There is a distinct cluster of positive residuals (model underestimates) over all periods for the three sites at the lowest elevations (sites 1, 12, and 15). As seen in Figure 3 and previously discussed, sites at low elevation and very close to the coast are subject to exceptionally high dry deposition, which does not conform to the patterns seen over the further inland and higher elevation areas. Including

these outliers in the regression led to a linear trend between residuals and Elevation/Distance from coast at the other sites, and hence a sub-optimal R^2 value. It is recommended that a spatial or spatial–temporal model of chloride deposition should omit sites that are very near the coast and at low elevation (in this case, this may be within 100 m from the cast and below 50 m ASL) or include a suitable non-linear component to allow for sea-spray effects if sufficient data are available.

5.4 | Potential applications and opportunity for complementary research

The findings of this paper are important for hydrological studies (e.g., groundwater recharge estimation, catchment water balance), corrosion predictions and ecological studies in Norfolk Island and other small islands elsewhere, which are typically characterized by a lack of data and limited monitoring options. Further investigations in Norfolk Island are underway to understand the effect of vegetation on atmospheric chloride deposition rates, considering that chloride deposition has been found to be higher under vegetation when compared to open areas (Eriksson, 1959; Ulrich, 1983). This will allow for a more complete understanding of spatio-temporal chloride deposition across Norfolk Island (and other small islands) as well as a historical chloride deposition in Norfolk Island to be estimated considering both spatial and temporal factors.

To further investigate the temporal variability of chloride deposition, the use of more detailed weather data, in combination with rainwater sample collection in shorter sampling periods would be beneficial. Examples of monitoring systems for finer time intervals include the use of autosamplers or self-logging tipping buckets that measure both rainfall volume and electrical conductivity automatically. Some additional factors to be further investigated include rainfall intensity (e.g., mm. 30 min^{-1}), local wind speed (e.g., using weather radar) and wave height. This level of detailed data requires a higher investment of resources in fieldwork and the data collection process.

5.5 | Chloride deposition estimates across small islands globally

The results of this study were compared with previous bulk chloride deposition estimates in small islands (Figure 8; Table 1). Figure 8 displays chloride deposition observations with respect to distance from

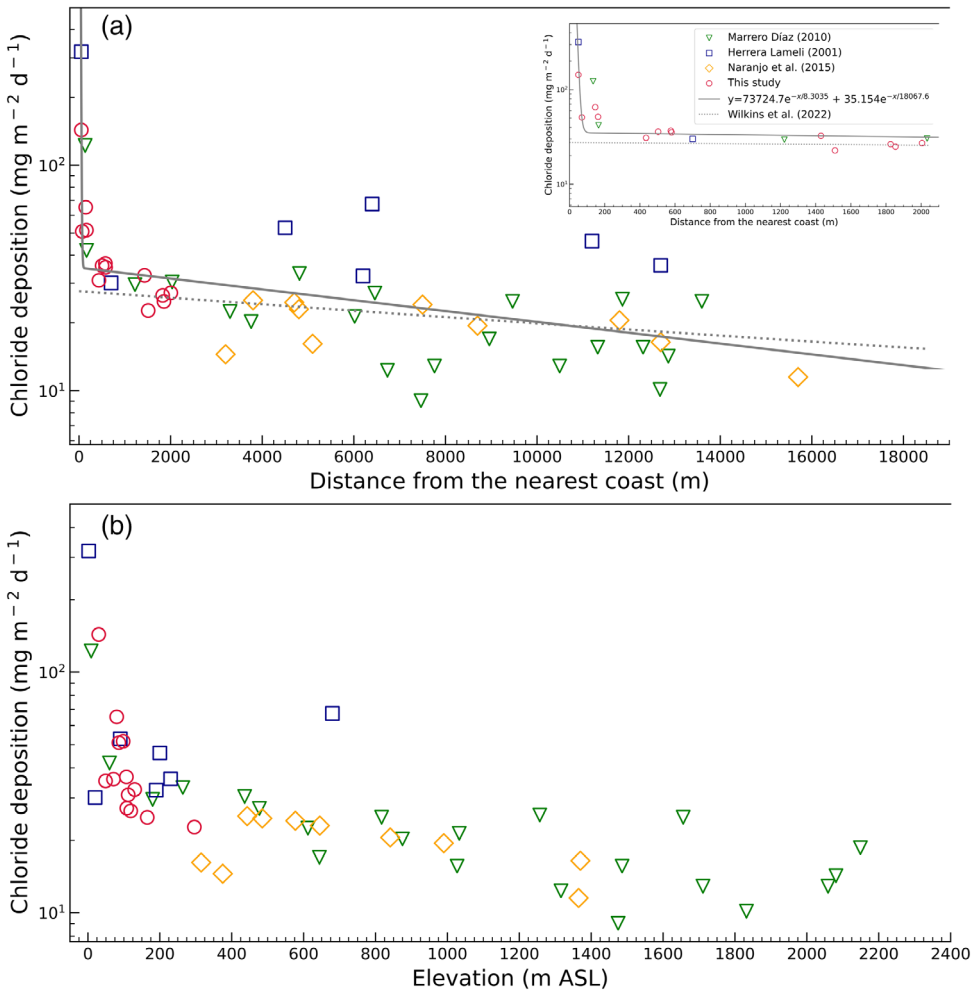

FIGURE 8 Chloride deposition estimates in small islands. The figure depicts chloride deposition measurements in small islands in respect to distance from coast and elevation, integrating data from both the current study and previous observations in small islands. Additionally, the figure displays the small islands-specific double exponential decay model and the continental model by Wilkins et al (2022) for comparision.

the coast and elevation, integrating measurements from this study with observations from other small islands. Only studies that used continually open collectors were included, meaning that studies that estimated either only dry or wet deposition, as well as studies that used a very distinct method (e.g., rainwater tanks) were excluded. Estimates for small islands situated close to continents were also excluded.

The chloride bulk deposition values quantified our Norfolk Island study generally agree with those from the Canary Islands (Herrera Lameli, 2001; Marrero Díaz, 2010; Naranjo et al., 2015) (Figure 8). The higher chloride deposition values in Fuerteventura Island (Herrera Lameli, 2001) in the range of 5–12.5 km from the coast (Figure 8a) can be attributed to lower elevations at these sites (Figure 8b). In the range of 500–2000 m from the coast, the results of our Norfolk Island study present similar values to those of Herrera Lameli (2001) and Marrero Díaz, 2010). For sites located up to 200 m from the coast, the results were distinctly different between studies, and between sites in the Norfolk Island study. This variation is expected since proximity to the ocean can lead to higher variations of chloride deposition, depending not only on the distance from the coast but also on differences in elevation, wind direction and average wind speed/wind gust.

The combined small island results show that distance from the coast and elevation present a significant relationship with deposition (Figure 8), while no significant relationship was found between spatial variations of rainfall and deposition. Figure 8 shows that there is a fast decrease of chloride deposition within around 200 m from the coast.

Previous continental scale studies in Australia have derived a double exponential decay equation expressing the strong chloride deposition dependence on distance from the coast (Hutton, 1976; Keywood et al., 1997; Wilkins et al., 2022). Keywood et al. (1997) applied a double exponential decay regression to explain the relationship with distance from the coast, representing a fast chloride deposition decay in the first 120 km from the coast due to aerosol deposition and a slow component beyond 120 km related to volatilisation of gaseous chlorine. Wilkins et al. (2022) presented an updated double exponential equation for Australia based on Keywood et al. (1997) model, using 367 previous chloride deposition measurements. Interestingly, the very fast chloride deposition decrease found in the small islands dataset is not predicted by the Wilkins et al. (2022) continental scale model (Figure 8a). This highlights the importance of chloride deposition models specifically designed for small islands or other areas close to the coast, especially considering the common lack of local data.

Following Keywood et al. (1997) and Wilkins et al. (2022), a double exponential decay regression was applied to small islands (Figure 8a) to express the relationship between distance from the coast and chloride deposition. The dataset comprises 52 observations, as depicted in Figure 8, combining findings from this study and previous observations for other small islands. Sites 12 and 15 were excluded from this data set due to their low number of samples and unrepresentative deposition values (see Figure 3, Table A1). To optimize the parameters of the double exponential model, the sum of squared errors between the log-transformed observed and modelled deposition values was used. The resulting double exponential model is:

$$D_{BP} = 73724.7e^{-\frac{x}{8.3035}} + 35.154e^{-\frac{x}{18067.6}}, \quad (3)$$

where D_{BP} is chloride deposition rate [mg m^{-2} d^{-1}] and x is the distance from the coast [m].

The model effectively explains the spatial variation of chloride deposition in these small islands, as evidenced by an R^2 value of 0.74. It indicates a very fast decay component within around 200 m from the coast, likely attributed to direct sea spray deposition, followed by a subsequent fast decay related to the settling of aerosol particles from the atmosphere (Figure 8a). A potential slow component due to gaseous chlorine is masked by the short distances from the coast in this data set. The presence of the very fast component, which is not present in the continental model, reinforces the need for specialized models to capture the unique dynamics in small islands.

It is worth noting this double exponential model for small islands does not consider temporal variations of chloride and the observed data in Figure 8a may not represent long-term deposition at each location. Although this limits the precision of the proposed model, it is still useful for small islands and coastal areas worldwide where there is a lack of locally-collected data and previously suggested models are not representative. To enhance our comprehension of spatial variability in chloride deposition within and between small islands at a global scale, future studies could incorporate long-term measurements at each location. Characterizing the effects of rainfall and wind speed, which were shown to be important in the present study, may further improve the generalized model. Additionally, it may be valuable to consider other spatial variables, such as elevation, up-wind distance from coast in the prevailing wind direction and spatial differences in wind speed.

6 | CONCLUSIONS

Understanding the variability of chloride deposition is crucial for hydrological investigations, particularly in small islands and coastal areas where chloride deposition can fluctuate greatly. This study contributes to knowledge of chloride deposition variability in small islands and coastal areas with similar characteristics by comprehending the influence and relative importance of spatial and temporal factors on chloride deposition in a small island, using statistical and regression analysis. To the best of the authors' knowledge, this is the first paper combining a high density of monitoring sites (15 sites/35.7 km^2) on a small island, a monitoring period of 23 months, and statistical modelling of factors controlling the spatial and temporal distribution of chloride deposition. Furthermore, a double exponential model for spatial distribution of chloride deposition in small islands worldwide is presented.

Chloride deposition rates in Norfolk Island presented high variation in space and time. Total deposition rates for the period September 2020 to July 2022 varied from 22.7 and 36.7 mg m^{-2} d^{-1}

for sites located more than 400 m from the coast. A higher variation was observed closer to the coast, with values varying from 50.9 to 1022.7 mg m^{-2} d^{-1} for sites located between 50 and 200 m from the coast. The data analysis reveals that variations in rainfall and wind gust over time significantly contribute to chloride deposition temporal variations, while distance from the coast and elevation influence the spatial variations of chloride deposition. Additionally, the results indicate that the prevailing wind direction can also influence the spatial distribution of chloride deposition.

Multiple regression analysis, including both spatial and temporal factors, demonstrates that rainfall (log-transformed period-total rainfall depth), wind gust (period-average maximum daily wind gust speed), distance from coast (distance from the nearest point on the coast) and elevation (elevation above mean sea level) explain 57% of log-transformed chloride deposition variability in Norfolk Island. The standardized regression coefficients indicate rainfall and wind gust have very similar effects on the temporal distribution of chloride deposition, while distance from coast has a more definite effect than elevation for its spatial distribution.

Interestingly, the results indicate that temporal effects influence chloride deposition differently depending on location. Temporal regression applied at each site separately shows that rainfall has a more definite effect when compared to wind gust for sites located in the center of the island. Conversely, for sites near the coast, wind gust emerges as the dominant factor in temporal effects.

The effect of distance from coast and elevation observed in Norfolk Island concurs with international studies in small islands. Previous continental-scale models that predict chloride deposition based on distance from the coast, as developed for Australia, do not capture the rapid decline of chloride deposition observed in small islands. This underscores the need for dedicated modelling for small islands. In addition to the model introduced for Norfolk Island, this study proposes a double exponential model for estimating chloride depositon in small islands using globally published data, representing a valuable tool for regions with limited local data. Refining this small island model involves the consideration of additional factors controlling chloride deposition, building on insights from the Norfolk Island study.

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DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are available in the supplementary material of this article.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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