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A qualitative investigation into scavenging of
airborne sea salt over Macquarie Island

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A QUALITATIVE INVESTIGATION INTO SCAVENGING OF AIRBORNE
SEA SALT OVER MACQUARIE ISLAND

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ABSTRACT

A comparative study is made on the amount of sea salt (dominantly NaCl) deposited on Macquarie Island due to atmospheric precipitation. It is found that the scavenging of solid salt particles alone cannot account for all the salt budget over certain areas of the Island. It is considered that sea spray droplets carried aloft by winds and scavenged by precipitation in the immediate vicinity of the shoreline is responsible for this deficit.

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1.   INTRODUCTION

A relatively stationary concentration of water droplets, as in clouds, can capture aerosol particles (AP) by simple Brownian diffusion.   Some of these droplets may subsequently fall to earth as precipitation.   Four attachment processes are known to remove aerosol particles from the atmosphere; convective diffusion; thermophoresis and diffusiophoresis; turbulent shear and turbulent inertial capture; and gravitational or inertial capture (Pruppacher and Klett 1980).   Of the four removal mechanisms only the first and last will be considered as being relevant in the ensuing discussion.   This allows a first-order treatment of the problem to be attempted.

The qualitative determination of salt (NaCl dominant) deposited on subantarctic islands is important in ecological studies. Flora and fauna finely coexist and must constantly battle the elements in order to survive (Jenkin 1972).   In addition to extremes of climate, subantarctic islands are subject to deposition of airborne sea salt which is leached into the soil (Jenkin 1972; Evans 1970). Soil nutrients are of importance to the productivity of vegetation and to omnivore/ herbivore populations.

Attachment or scavenging and removal mechanisms can be classified in two ways; particles can be taken from the atmosphere by precipitation (e.g. rain, snow, hail, settling fog droplets) or removed whilst in a particular state.   These processes are referred to as "wet" and "dry" removal processes respectively. Removal processes are difficult to quantify even in specific cases and drastic simplifications and parameterisations need to be made in order to make problems more tractable. This report gives a simplified treatment best described as a first-order analysis.

2.   THEORY

A brief outline into the theory of scavenging by convective diffusion of aerosol particles by cloud droplets is given before commencing with a discussion of the problem.

Let $n(\underline{r}\ t)$ denote the concentration of diffusion aerosol particles at $\underline{r}$ at a time $\underline{t}$. The current density in a stationary medium is given by

$$\underline{J} = - D\underline{\nabla}n \tag{1}$$

where D is the diffusion coefficient.   From the continuity equation we have

$$\frac{\partial n}{\partial t} = - \nabla.\underline{J} \tag{2}$$

Substituting equations (1) into (2) for $\underline{J}$ gives

$$\frac{\partial n}{\partial t} = D\nabla^2 n \tag{3}$$

where D has been taken to be constant.   Equation (3) is known as the diffusion equation.

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The mean square displacement of a distribution of diffusing particles is given by

$$\int_{-\infty}^{+\infty} z^2 \frac{\partial n}{\partial t} dz = N \frac{\partial \langle z^2 \rangle}{\partial t} = D \int_{-\infty}^{+\infty} z^2 \frac{\partial^2 n}{\partial z^2} dz = 2DN \qquad (4)$$

where N is the number of identical aerosol particles per unit area introduced at time $t = 0$ in an infinitesimally thin slab near $z = 0$.
Hence, the mean square displacement becomes

$$\langle z^2 \rangle = 2Dt \qquad (5)$$

Einstein (1905) derived $\langle z^2 \rangle$ somewhat differently and found that

$$\langle z^2 \rangle = \frac{kTt}{3\pi\eta_a r} \qquad (6)$$

where k = Boltzmann's constant
T = absolute temperture (degrees Kelvin)
η_a = dynamic viscosity of air
r = radial distance of spherical particle
t = time.

η_a is related to temperature by

$$\eta_a = (1.718 + .0049T) \times 10^{-4}, \qquad\qquad T(^oC) \geqq 0^oC$$

$$\eta_a = (1.718 + .0049T - 1.2 \times 10^{-5}T^2) \times 10^{-4}, \quad T(^oC) < 0^oC \qquad (7)$$

Equating equations (5) and (6) yields

$$D = \frac{kT}{6\pi\eta_a r} \qquad (8)$$

Cunningham (1910) found experimentally that a more suitable form of the diffusion coefficient for small particles can be given by

$$D = \frac{kT(1 + \alpha N_k)}{6\pi\eta_a r} \qquad (9)$$

where $\alpha = 1.257 + .4\exp(-1.10/N_k)$

N_k is the Knudsen number and is found from $N_k = \lambda_a/r$ where λ_a is the mean path of free air molecules and r_a is the characteristic dimension of a particle. λ_a is given by

$$\lambda_a = \lambda_o \left[\frac{p_o}{p} \right] \left[\frac{T}{T_o} \right] \qquad (10)$$

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where $\lambda_O = 6.6 \times 10^{-6}$ cm

$p_O = 1013.25$ mb

$T_O = 293.15$ ^OK

p = atmospheric pressure.

The loss rate of aerosol particles per unit volume of air due to scavenging by cloud drops is given by

$$\frac{\partial n(\underline{r}, t)}{\partial t} = n(\underline{r}, t) \int_0^\infty K(\underline{r}, a)n_d(a, t) \, da$$

(11)

where a denotes raindrop radius, $K(\underline{r}, a)$ is the collection kernal and $n_d(a, t)$ is the number of drops per unit volume of air at time t in a size interval a to a + da. The scavenging coefficient Λ is derived from equation (11). Λ can be defined as the fractional depletion rate of aerosol concentration where

$$\Lambda(\underline{r}, t) = \int_0^\infty K(\underline{r}, a)n_d(a, t) \, da$$

(12)

Following the method adopted by Pruppacher and Klett (1980) Λ can be expressed as

$$\Lambda = 4\pi D \int_0^\infty a \, n_d(a) \, da \approx \frac{1.35 w_L D}{\bar{a}^2}$$

(13)

where $n_d(a)$ da is the number of drops per cm^3 in the radius range (a, a + da), w_L is the liquid water content of the cloud in gcm^{-3} and $\bar{a}$ is the average drop radius in cm.

If $n_d(\underline{r}, t)$ does not vary with time then Λ becomes constant so that

$$n_d(\underline{r}, t) = n_d(\underline{r}, o)\exp(-\Lambda t)$$

(14)

From equation (14) the half-life $t_{\frac{1}{2}}$ of aerosol particles can be shown to be

$$t_{\frac{1}{2}} \approx \frac{(\ln 2) \, \bar{a}^2}{1.35 w_L D}$$

(15)

Thus, given a reasonable estimate for $\bar{a}$, w_L and D for given atmospheric conditions, Λ can be calculated and used to find a theoretical qualitative estimate of the amount of aerosol particles deposited in precipitation. This problem is dealt with in subsequent sections.
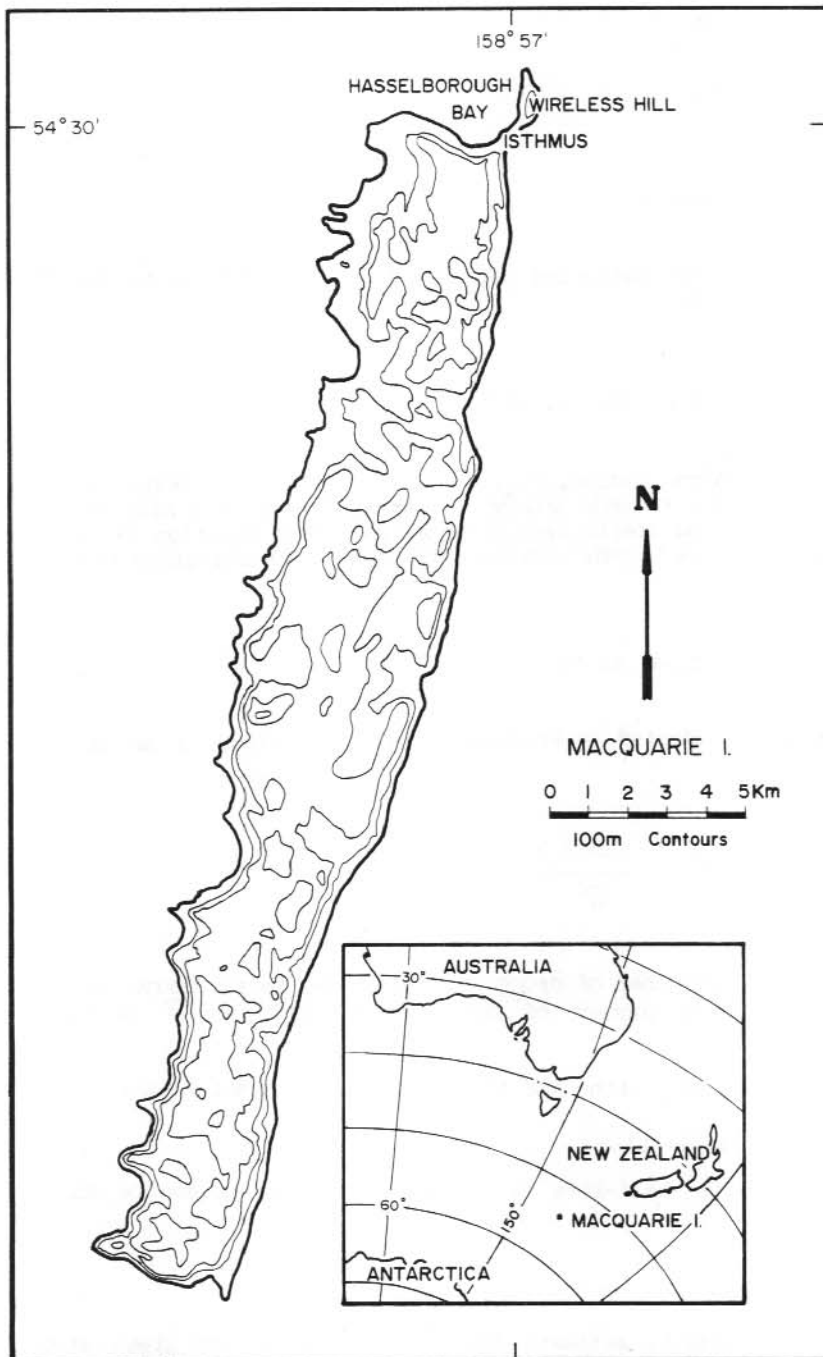
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Figure 1. Contour map of Macquarie Island.

3. ENVIRONMENT

Macquarie Island (54°38'S, 158°53'E) is a small isolated subantarctic island situated in the Southern Ocean some 1600 km south-east of Tasmania, Australia and 1530 km north of the Antarctic coastline. It is a long narrow plateau rising on average 250-350 m above sea level and bounded mostly by cliffs or steep slopes (Figure 1). The Island has an area approximately 120 km² and is some 34 km long and from 2.5 to 5 km wide.

Macquarie Island has one of the world's most uniform climates (Landsburg et al. 1965). The mean annual air temperature at ground level is 4.7°C with a mean annual and mean diurnal variation of $\pm$ 3.3°C and $\pm$ 1.6°C respectively. The mean relative humidity is 89% and precipitation occurs on an average of 312 days of the year. The mean wind velocity is 8.6 m/s with an average cloud cover of 83%. The low cloud ceiling is some 500-700 m on days when precipitation occurs. The climate is dominantly oceanic.

A chemical analysis of precipitation at The Isthmus, Wireless Hill and on the plateau was undertaken by Evans (1970) and is reproduced in Table 1.

Site details	Isthmus	Wireless Hill	Plateau
Altitude (m)	6	100	235
Distance from west coast (m)	35	150	2500
Mean annual precipitation (mm)	926	926	1074
Chemical component	($kg\ ha^{-1}yr^{-1}$)		
Cations:			
Na	9556.32	2870.60	209.43
K	951.93	222.24	9.67
Ca	416.70	125.94	17.18
Mg	1228.80	387.07	41.89
Anions:			
Cl	17501.40	5278.20	418.86
SO_4	536.31	768.58	88.07
HCO_3	248.17	ND	ND
Colloidal matter	653.76	ND	ND
Other solids	10.46	4.26	4.30
Total solids	33104.50	9815.60	784.02

ND = not determined

Table 1. Chemical analysis of precipitation at The Isthmus, Wireless Hill and the plateau.

The major constituent of ions in freshwater lakes on Macquarie Island was found to be Na and Cl. Evans (1970) suggested that the surrounding ocean is responsible for these dissolved salts. This is supported by the ionic ratios for these contained waterbodies which are similar in composition to the surrounding sea water. Soluble material in lake water is marine derived, most likely carried inland by windborne sea spray and/or atmospheric precipitation.

Precipitation at Macquarie Island is generally relatively short-lived and heavy (showers) or long-lived and light (drizzle). From the author's experience the frequency of both types of precipitation is fairly even. The Island's meteorological records for the period 1948 to 1982 verify this observation.*

Table 2 lists a range of relevant parameters which are representative of average cloud conditions for a maritime environment including the mean sea salt particle (NaCl) radial size r (Pruppacher and Klett 1980).

T (°C)	p (mb)	r (cm) 10^{-4}	a (cm) 10^{-4}	w_L (gcm^{-3}) 10^{-6}
−5 − 0 (−3)	800 − 900 (850)	.1 − .3 (.25)	5 − 10 (8)	.4 − .8 (.6)

Table 2. Relevant average water droplet and cloud parameters for a maritime environment. The variables T, p, a and w_L represent the atmospheric temperature and pressure, radial cloud droplet size and cloud water content at an altitude of 500−900 m respectively. Brackets indicate mean values used in this analysis.

Bubble size (diameter) (μm)	Mass of salt particle (g)	Dry salt particle radius (μm)
2000	1.5×10^{-7}	25
100	1.9×10^{-11}	1.3
20	1.5×10^{-13}	.3

Table 3. Bubble size related to dry salt particle radius produced.

*Meteorological data on Macquarie Island was obtained from Evans (1970) and the Bureau of Meteorology, 150 Swanston Street, Melbourne, Victoria, Australia, 3000

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4. FORMATION OF AIRBORNE SEA SALT PARTICLES

Airborne sea salt is formed by the bursting of air bubbles produced by the entrapment of air at wave crests, and of less importance, as a result of wind action spray drops at the crest of surface waves. See Woodcock et al. (1953), Kientzler et al. (1954), Knelman et al. (1954), Mason (1954b), Moore and Mason, (1954) and Blanchard, (1954) for studies done in this field.

As an air bubble rises to the suface of the ocean it thins out on top and bursts (Figure 2a). Fragments of the burst water film are thrown upwards by the out rushing air (Figure 2b). A narrow jet will be formed as a result of water filling the bubbles (Figure 2c). This jet eventually breaks due to instabilities and forms a few large and several smaller drops (Figure 2d). Depending on the relative humidity and air turbulance some drops fall to the ocean surface. The remainder evaporate whilst airborne, leaving a small dry sea salt particle light enough to be carried aloft by air currents.

Table 3 summarises data relating bubble size to dry salt particle radius produced.
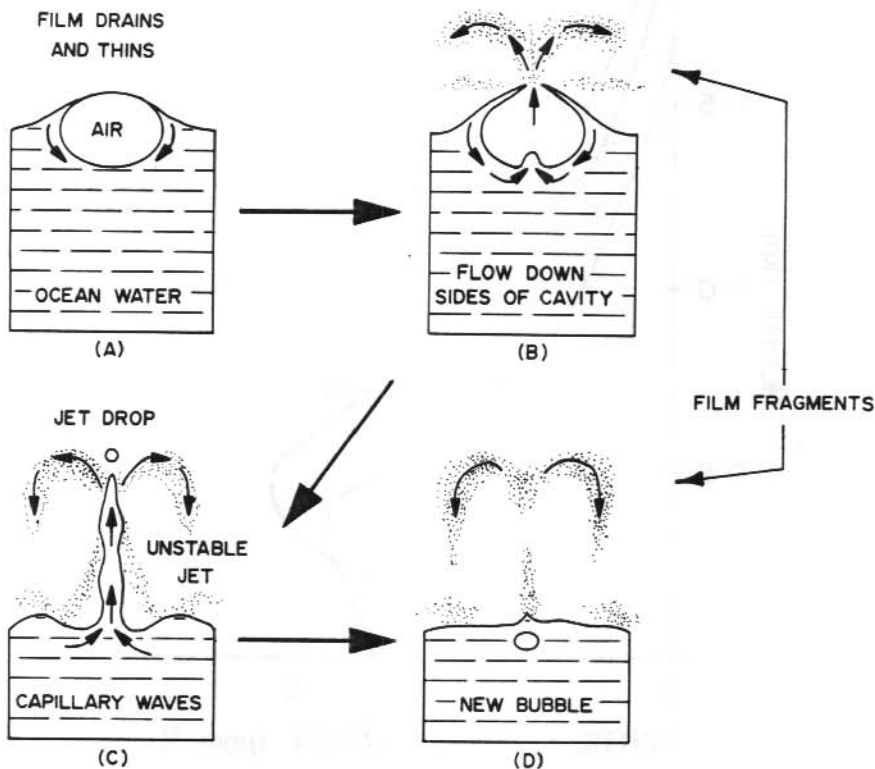

Figure 2. The four stages of the formation of sea salt particles by the bubble-burst mechanism. (From Day (1965) with changes.

The bursting of the bubble also produces large numbers of small dry salt particles. Mason (1954b and 1957b) found that salt particles liberated this way have masses less than 2×10^{-14}g equivalent to a dry radius of about .1 µm. The largest and smallest particle masses vary from 2×10^{-13} g to 10^{-15} g corresponding to a dry radius of .3 to .07 µm respectively. The average air-borne salt mass is about 3×10^{-14} g corresponding to a dry radius of 25 µm.

Salt profile size is found to increase from sea level to about 500 m above when an exponential decrease is observed to an elevation 2 to 3 km above sea level (Figure 3). Plot (1) was obtained over the Pacific near Hawaii, plot (2) over the Atlantic near Florida and plot (3) over the Carribbean (Erikson, 1959). No data is available for the Southern Ocean, but for the purposes of this investigation plots (1) and (2) will be taken to be representative of the subantarctic region.
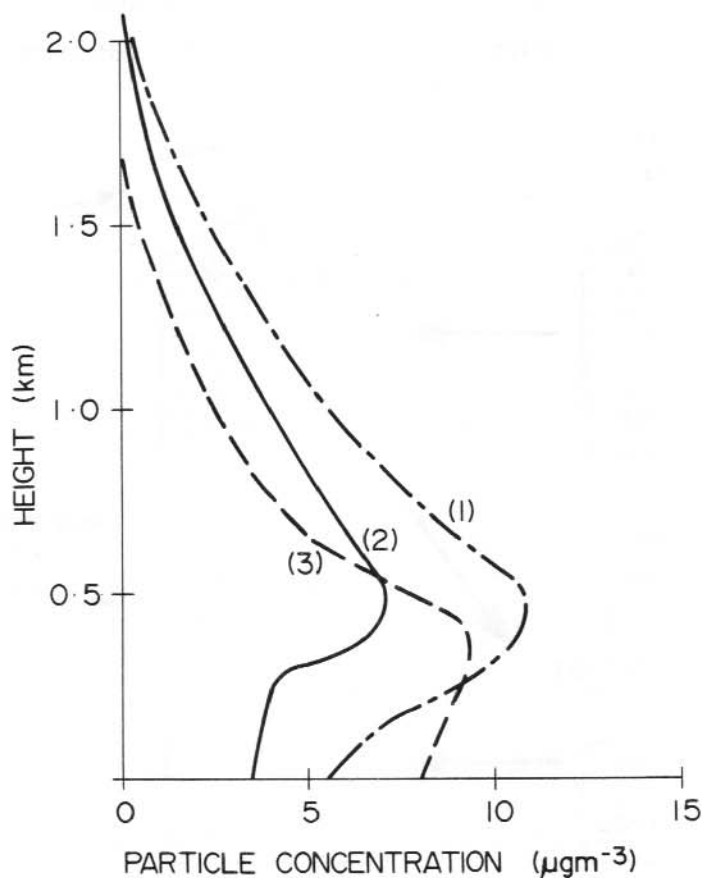

Figure 3. Vertical variation of sea salt concentration in air over (1) the Pacific near Hawaii, (2) the Atlantic near Florida, and (3), the Caribbean. (From Erikson (1959) based on data from Woodcock (1953)).

5. SCAVENGING OF AIRBORNE SALT

Using data from Table 2 the following quantities are determined:

$\lambda_a = 7.25 \times 10^{-6}$, $N_k = .290$, $a = 1.266$, $\eta_a = 1.7 \times 10^{-4}$ and from equations (9) and (13) $D = 6.36 \times 10^{-7}$ and $\Lambda = 8.1 \times 10^{-7}$ respectively.

Assuming that the total salt particle mass concentration at 1000 m is 4-6 μgm^{-3} (Pruppacher and Klett, 1980) for wind speeds in the order of 8 m/s, particle concentration can be calculated. This is derived by dividing the particle concentration at 1000 m by the average airborne mass of dry salt which is about $3 \times 10^{-14}g$ (Erikson 1959; Peterson and Junge 1971). Salt particle concentration is calculated at 133 to 200 cm^{-3} with a mean of 167 cm^{-3}. Hence, $\Lambda nt_{\frac{1}{2}}$ particles are absorbed per cm^2 of cloud air and $\Lambda nt_{\frac{1}{2}} w_L^{-1}$ particles are absorbed per cm^3 of cloud water. It is readily shown that $\frac{1}{2} \Lambda nt_{\frac{1}{2}}$ = 116 cm^{-3} and $\Lambda nt_{\frac{1}{2}} w_L^{-1}$ = 1.93 $\times 10^8$ cm^{-3} which corresponds to

$$\Lambda nt_{\frac{1}{2}} w_L^{-1} \times 10^8 \ cm^2/ha \times 10^2 \ cm/yr \times 3 \times 10^{-17} \ kg = 58 \ kg/ha/yr$$

of salt (NaCl) to fall as a result of in-cloud scavenging over Macquarie Island. No account is made of below-cloud scavenging at this stage.

Evans calculated from _in situ_ sampling that NaCl falls on the plateau, Wireless Hill and The Isthmus at a rate of 650, 3700 and 23 000 kg/ha/yr respectively. These figures are 11.2, 63.8 and 397 times greater than the 58.0 kg/hr/yr figure calculated above. This suggests that "in cloud" scavenging as a result of convective diffusion cannot alone account for the total salt deposition on Macquarie Island. The salt differences can only be accounted for by below-cloud scavenging.

A less rigorous solution to this problem is possible which considers the effects of forced convective diffusion, wind turbulence, shear diffusion and inertial impaction. Since approximate descriptions exist for most of these processes only a rough estimate can be made. In- and below-cloud scavenging is implicitly included in the analysis. This method is discussed by Pruppacher and Klett (1980). Briefly, the problem is to obtain a reasonable upper bound Λ_M for the nett scavenging coefficient in a precipitating cloud. Case studies by Slinn and Hales (1971), Dingle and Lee (1973), Crandall et al. (1973), Young (1974) and Williams (1974) imply that in-cloud scavenging by convective, precipitating systems can be regarded as a two-stage process. First, salt particles are absorbed by cloud water through various scavenging mechanisms (rainout) such as nucleation, Brownian and turbulent diffusion, and diffusio- and thermophoresis. Secondly, cloud water itself is scavenging primarily through inertial capture by precipitating cloud particles (washout) such as rain, snow and hail. Pruppacher and Klett suggest that Λ_M can be given by

$$\Lambda_M (sec^{-1}) \simeq 4.2 \times 10^{-4} \overline{E} \ R^{0.79} \tag{16}$$

where $\overline{E}$ is the characteristic collision frequency for the drop-droplet interactions and R is the rain rate in $mmhr^{-1}$. $\overline{E}$ is taken to equal .83 from Crandall et al. (1973). Λ_M has been calculated for various rain rates R. On average it rains some 1000 mm per 312 days of the year at Macquarie Island. This corresponds to R = .134 $mmhr^{-1}$ for 312 days of continuous precipitation. However atmospheric precipitation is not continuous and on

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average precipitation is recorded from 3 to 8 hours per day. This means that R will vary from .401 to 1.07 mmhr^{-1} per day for the 312 days of precipitation. From equation (16) it is readily seen that as R varies so will Λ_M. Table 4 summarises the amount of NaCl deposited (Q_M) in a time interval $t_{\frac{1}{2}}$ as a function of R.

Table 4 is calculated for n = 167 cm^{-3} and assuming an average NaCl mass of 3 x 10^{-14} g. Variations in n and particle mass will change Q_M and so values of Q_M should only be regarded as representative. The figures in the fourth column of Table 4 represent the half-life of NaCl capture during in- and below-cloud scavenging. Dividing $t_{\frac{1}{2}}$ into the time period of continuously recorded rain in hours (figures in brackets of column one) indicates the number of times the atmosphere is flushed of all aerosol particles during the interval of precipitation. Sellers (1965) has estimated that for oceanic areas with an average rainfall of 112 cm per year and with good scavenging efficiency the troposphere could be cleansed of aerosol particles as much as a thousand times a year. For example if it rains an average of 4 hours per 312 days then the atmosphere could theoretically be cleansed about 1000 times per year.

R (mmhr^{-1})		Number of equivalent days of continuous precipitation	Λ_M sec^{-1} x 10^{-5}	$t_{\frac{1}{2}}$ hr		Q_M kg/ha/yr
.134	(24)	312	7.1	2.7	(8.9)	515
.401	(8)	104	17	1.1	(7.1)	411
.536	(6)	78	21	.92	(6.5)	376
.804	(4)	52	29	.66	(6.0)	347
1.07	(3)	39	37	.52	(5.8)	336

Table 4. NaCl deposited in a time interval $t_{\frac{1}{2}}$ as a function of R.

Site	Dry salt particles	Sea spray droplets	Total fallout
Plateau	350	0	650
Wireless Hill	450	3250	3700
Isthmus	525	22475	23000

Table 5. NaCl (kg/ha/yr) scavenged from the atmosphere due to dry salt particles and sea spray droplets.
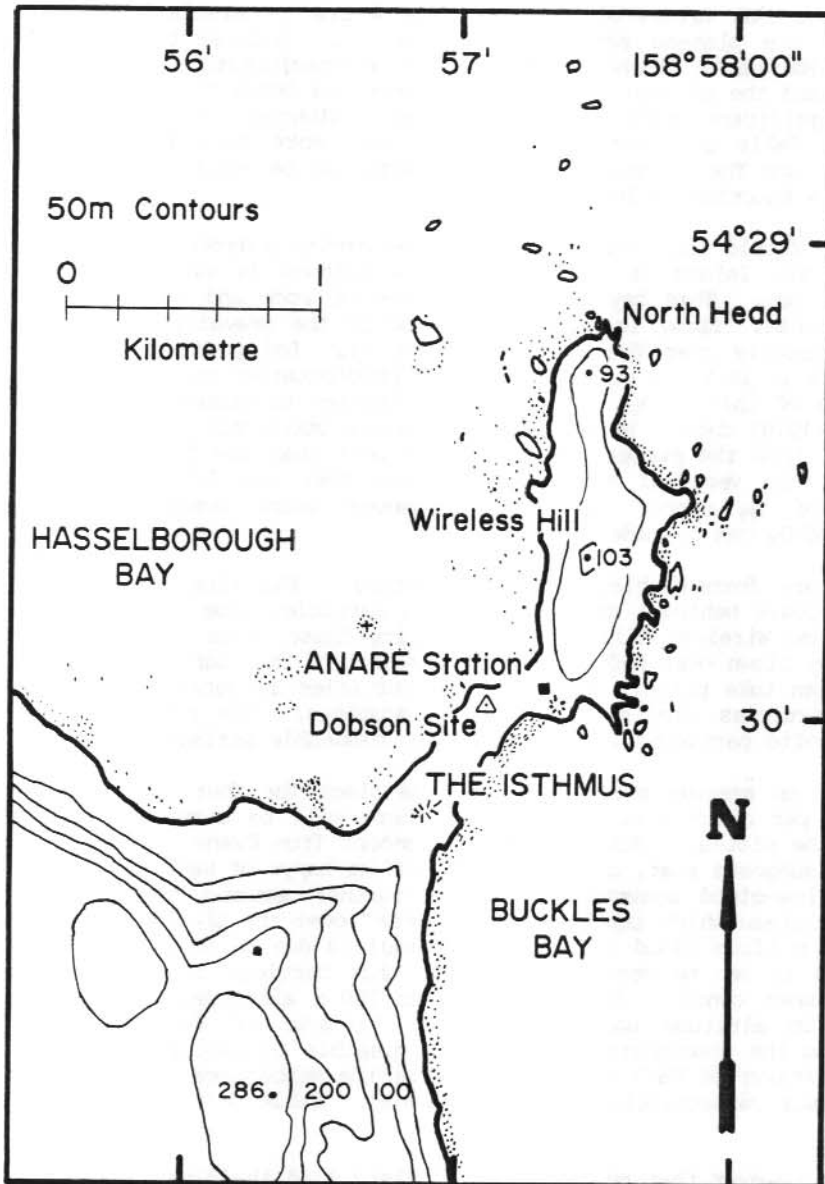
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Figure 4.   The Macquarie Island isthmus environment.

6.   DISCUSSION

It can be seen that values of Q_M from Table 4 are comparable with that found by Evans for the plateau region (650 kg/ha/yr). Because the average cloud ceiling over Macquarie Island during periods of precipitation is approximately 500 to 900 m and the plateau is some 300 m above sea level there is not expected to be any significant below-cloud scavenging. Although the same quantity of precipitation falls over the entire Island much more NaCl is deposited over Wireless Hill and The Isthmus. This is thought to be entirely due to washout processes as a function of local geography.

Why more NaCl should fall out this way can be better understood when the local geography of the Island is considered. The Isthmus is situated adjacent to Hasselborough Bay. This bay has an abundance of rock and reef outcrops over which waves break. Spray is carried downwind by the prevailing westerly winds which blow directly over The Isthmus (Figure 4). Being situated so close to salt laden air it is to be expected that any precipitation over The Isthmus will scavenge some of this spray. There are no figures to support this view other than Evans' (1970) data. Wireless Hill receives about six times more NaCl in precipitation than the plateau and six times less than The Isthmus. Without a knowledge of the vertical spray distribution over the Island a theoretical calculation of Q_M cannot be made. However using Evans' data a rough calculation of Q_M can be made.

Spray drops are formed chiefly by wave action. The finer drops eventually evaporate to leave behind a small salt (NaCl) particle. Due to the proximity of The Isthmus and Wireless Hill to the shoreline these sites are more likely to have sea spray blown over before much if any, due to the high relative humidity, evaporation can take place. Hence aerosol particles in general are made up of both solid particles (dry salt) and spray droplets. The proportion of spray droplets to solid particles is unknown but a reasonable estimate can be derived.

Assuming that on average precipitation takes place for four hours per day for the 312 days per annum that rain occurs, there will be a maximum fall of 350 kg/ha/yr on the plateau. Subtracting this amount from Evans' derived figure of 650 kg/ha/yr suggests that, at most, about 300 kg/ha/yr of NaCl is captured as a result of below-cloud scavenging. It is further assumed that no sea spray reaches the plateau which implies that "washout" over the plateau for an average height of 400 m (from cloud ceiling to plateau) is due to solid salt particles. From Figure 3 it can be reasonably assumed that particle concentration can be considered almost constant from sea level to 700 m altitude. Hence for every 400 m drop in altitude approximately 300 kg/ha/yr of dry salt (NaCl) is scavenged from the atmosphere. It is then possible to calculate 3250 kg/ha/yr and 22 475 kg/ha/yr of NaCl is scavenged from the atmosphere over Wireless Hill and The Isthmus respectively due to sea spray. Table 5 summarises the above data.

It is assumed however that no sea spray droplets find their way to the plateau. Even if a small proportion of spray droplets did it would not alter the finding that the greatest scavenging of spray droplets occurs on The Isthmus and probably the whole west coastline.

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7. CONCLUSION

It is concluded that scavenging of solid salt (NaCl) particles due to precipitation cannot alone account for the total amount of salt that is deposited over some of Macquarie Island. The difference is made up by the scavenging of sea spray droplets which has more significance with proximity to the shoreline.

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