

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

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(with ten tables and two text-figures)

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Through study of rainwater collections made at Macquarie Island, during the 1985-86 austral summer, it was possible to compare previous theoretical predictions of sea-salt scavenging in precipitation with actual observations. Some earlier rainwater collections are found to have grossly overestimated the initial salt budget. It is shown that the earlier theoretical investigation adequately accounts for the salt budget and that sea-spray droplets play little part in salt scavenging by precipitation over most parts of the island.

Key Words: sea salt, salt scavenging, Macquarie Island.

INTRODUCTION

Calculations, based on actual rainwater measurements obtained from Evans (1970) and Jenkin (1972), led to the conclusion that in- and below-cloud scavenging processes, due to solid sea-salt particles, could only account for part of the observed total salt (NaCl) budget over Macquarie Island, the balance being due to sea-spray droplets (Mallis 1985). Subsequently, rainwater collections were performed, from December 1985 to April 1986 inclusive, for various sites over the island. Chemical analysis of chlorides and sulphates tends to support previous theoretical predictions (Mallis 1985) and reject Evans' data; Jenkin's data are also found to be suspect, though to a lesser degree. It can be demonstrated that it is possible to account for the expected salt budget without having to make up the difference by resorting to sea spray.

Even though drastic simplifications and parameterisations have been made in order to simplify the theory, they do make problems more tractable. This report follows up earlier work in this field and provides quantitative data to support previous theoretical predictions.

PROCEDURE

The experimental procedure for collecting water samples is described below. In all, four sites were chosen as representative of anticipated different conditions to be encountered over the island

and within easy access of the Australian National Antarctic Research Expedition (ANARE) station. Table 1 lists these sites, together with their approximate altitudes and commissioning dates. Site 1 was located in the Bureau of Meteorology

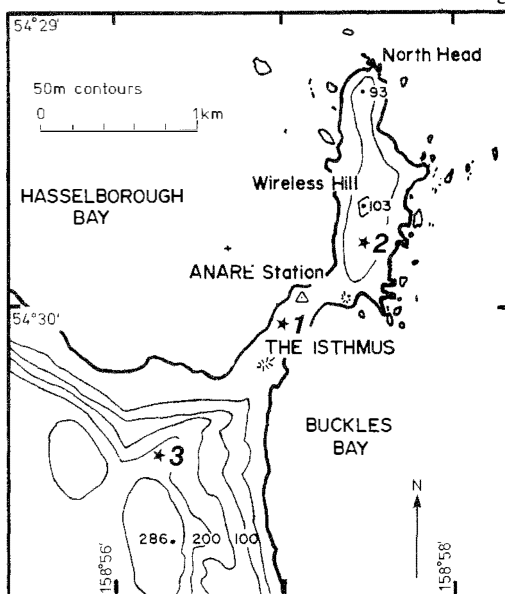


FIG. 1 — Map of the isthmus and its immediate environ, with approximate positions of rainwater collection sites 1, 2 and 3. Site 4 is located about 3 km south of site 3.

TABLE 1
Water collection site details

Site name	Site number	Commissioning date	Altitude (m)
Isthmus	1	13 Dec 1985	4
Wireless Hill	2	18 Dec 1985	~100
Ski Hut	3	29 Dec 1985	~220
Mt Elder	4	23 Feb 1986	~300

Instrument Enclosure on the isthmus; site 2 was atop Wireless Hill, located near Lambing Gully, which also afforded some protection from direct wind gusts; site 3 was situated near North Mountain and close to the ski hut (see fig. 1). Site 4 was located on the plateau proper, at the base of Mt Elder on the western side. As only three collection containers were in use, sites 3 and 4 were alternated. Sites 2 and 3 approximate very closely to those originally chosen by Jenkin (1972); site 1 is, of course, identical. However, the sampling procedure adopted for this study differs from that used by Evans (1970) and Jenkin (1972) to the point where it is concluded that spurious results were originally obtained. This discrepancy is discussed further later.

Before each collection container (a plastic bucket with a lip diameter of 255 mm) was deployed in the field it received a thorough wash, first with tap and then with de-ionised water. In the field, each bucket was set into a solid styrene foam block which was firmly anchored to the soft soil. Rainwater was removed from these buckets only during times of no precipitation. All three would be attended to within one hour or, in the case of site 4, within two hours. Rainwater was decanted on-site into sterile 250 ml plastic bottles. About 50-100 ml of de-ionised water was used to wash the bucket

and also decanted, to help assure the integrity of the next water sample and keep a running check in the event of possible contamination arising through the water collection procedure. In order to arrest biological degradation which might otherwise alter the chemical composition, enough chloroform was added, in the laboratory, to each decanted water bottle, including wash water, to make up about a 10% solution.

Complete meteorological records were kept for the period of this study to help later correlation of rainfall events with subsequent salt concentrations for each site. These meteorological records are, strictly speaking, accurate only for site 1 but can be extrapolated to other sites with caution. Subsequent chemical analysis was performed by the CSIRO Division of Atmospheric Research in Melbourne.

In all, 263 250-ml sample bottles were collected, representing about 43 litres of rainwater over a four-month period. The success of this project depended upon obtaining enough water samples, covering as many individual precipitation events as possible, to perform subsequently a statistically relevant and meaningful analysis of the data. (Note that the chemical analysis accuracy is given as $\pm 3.5\%$ whilst rainwater volumes are accurate to ± 0.1 ml.)

TABLE 2
Monthly mean rainfall data for site 1

Month	Total rainfall	No. days rainfall	Approx. hrs rainfall	Average rainfall rate (mmhr ⁻¹)
Dec 1985	23.8	9	54	0.441
Jan 1986	73.0	25	210	0.348
Feb 1986	84.8	18	174	0.487
Mar 1986	136.2	28	357	0.382
Apr 1986	41.4	9	117	0.354

ANALYSIS

A full theoretical description of scavenging by convective diffusion of aerosol particles by cloud droplets is given in Mallis (1985) and will not be repeated here. However, the same mathematical symbols and definitions are retained.

A reasonable estimate of the scavenging coefficient, Λ , has been given by Pruppacher & Klett (1980), taking into account both in- and below-cloud scavenging. Λ is given by

$$\Lambda \approx 4.2 \times 10^{-4} \bar{E} R^{0.79} \text{ sec}^{-1}$$

where \bar{E} is the characteristic collision frequency for drop-droplet interactions and R is the rain rate in mmhr^{-1} . \bar{E} is taken to equal 0.83, from Crandall *et al.* (1973). A nestimate of R for site 1 is found from table 2, where the total rainfall rate is calculated as a monthly mean. The total mean rainfall rate over the five months was found to be $\bar{R} = 0.402 \text{ mmhr}^{-1}$, with a standard deviation of 0.060 mmhr^{-1} .

An estimate of the scavenged deposition rate, Q , of NaCl in kg/ha/yr is given by

$$Q = \Lambda n_x t_{1/2} W_L^{-1} A \quad (1)$$

where A is a conversion factor ($\text{kgcm}^3/\text{ha/yr}$), n_x is a mean NaCl particle concentration (cm^{-3}), $t_{1/2}$ is the half-life of NaCl particles and W_L is the liquid water content of cloud (gcm^{-3}). Best estimates of $t_{1/2}$, n_x and W_L for average conditions are $8.56 \times 10^5 \text{ sec}$, 167 cm^{-3} and $0.6 \times 10^{-6} \text{ gcm}^{-3}$ respectively. Only n_x is considered to vary significantly over the three collection sites, and so Λ , $t_{1/2}$ and W_L are considered constant in the ensuing analysis.

As determined earlier (Mallis 1985), it is possible to estimate the amount of in-cloud scavenging qualitatively, using equation (1). This was found to be about ($D^1 =$) 58 kg/ha/yr of NaCl deposited in precipitation. This value was estimated, using the above variable values of n_x , $t_{1/2}$ and W_L , and taking $R = R_x = 0.401 \text{ mmhr}^{-1}$. Note that R_x and \bar{R} compare extremely favourably. Also calculated was the total expected amount of NaCl deposited due to in- and below-cloud scavenging. This was found to be about ($Q_m =$) 411 kg/ha/yr for site 1. Having access to quantitative data, it is now possible to improve the original scavenging model and compare with actual observations as a test of its accuracy.

A new estimate of the actual in-cloud scavenging rate, D , can be given by

$$D = D'(\bar{n}_\lambda)/(\bar{n}_x) \quad (2)$$

where \bar{n}_λ is an estimate of the average NaCl particle concentration at cloud altitudes and in region A;

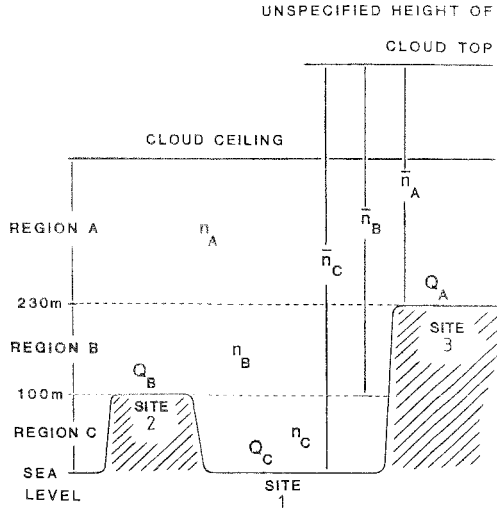


FIG. 2 — Schematic diagram of isthmus as viewed from west looking east, with regions A, B and C defined, along with relevant parameters.

similarly \bar{n}_B and \bar{n}_C are the averaged particle concentration values calculated within regions B and C, and regions A, B and C respectively (fig. 2 indicates these regions). These averaged particle concentrations are given by

$$\bar{n}_A = \bar{n}_x(P_A/Q_A) \quad (3)$$

$$\bar{n}_B = n_x[(P_B-D)/Q_B] \quad (4)$$

$$\bar{n}_C = n_x[(P_C-D)/Q_C] \quad (5)$$

where Q_A , Q_B and Q_C are the estimated total deposition rates due to scavenging of NaCl in precipitation within regions A, B and C respectively; also P_A , P_B and P_C are the actual deposition rates, as measured at the bottom of regions A, B and C respectively. P_A , P_B and P_C values are determined in the next section.

After making due allowance for differing rainfall rates within each region, Q_A , Q_B and Q_C can be estimated from

$$Q_A = Q_M(R_x/R_A)^{0.79} \quad (6)$$

$$Q_B = Q_M(R_x/R_B)^{0.79} \quad (7)$$

$$Q_C = Q_M(R_x/R_C)^{0.79} \quad (8)$$

where R_A , R_B and R_C are the estimated rainfall rates for regions A, B and C respectively. Table 3 lists five separate collections made at all sites. Total volume of rainwater plus the percentage difference between sites 2 and 3 with respect to site 1 is given.

TABLE 3
Volume of rainwater collected from all available sites at approximately the same time

Collection date/period*	Collection site	Volume rainwater collected (ml)	Percentage difference WRT site 1
29 Dec 1985	1	284.6	—
(2)	2	369.3	29.8
2 Jan 1986	1	381.6	—
(3)	2	476.0	24.7
	3	548.0	43.6
9 Jan 1986	1	167.2	—
(6)	2	233.4	39.6
	3	252.9	51.3
13 Jan 1986	1	159.1	—
(4)	2	170.9	7.4
	3	208.5	31.0
23 Feb 1986	1	3344.6	—
(12)	2	3728.9	11.5
	4	4305.6	17.2

* Numbers in parentheses indicate sample period, in days, over which collections were made.

These mean percentage differences of sites 2 and 3 are listed in table 4, along with their estimated mean rainfall rate. It is readily observed that $R_A > R_B > R_C$ on average.

Using the following equations, estimates of particle concentrations within regions A, B and C respectively are

$$\bar{n}_A = n_A \quad (9)$$

$$\bar{n}_B = [n_A(P_A - D) + n_B(P_B - P_A)] / P_B - D \quad (10)$$

$$\bar{n}_C = [n_A(P_A - D) + n_B(P_B - P_A) + n_C(P_C - P_B)] / P_C - D \quad (11)$$

Note that the RHS of equations (10) and (11) are, in fact, weighted means used in determining \bar{n}_B and \bar{n}_C . Equating equations (3), (4) and (5) with (9), (10) and (11) respectively, and substituting for Q_A , Q_B and Q_C from equations (6), (7) and (8) respectively, solutions for n_A , n_B and n_C can be found. Table 5 summarises these findings. Note that, although $n_C/n_A = 1/5.9$ and $n_B/n_A = 1/3.2$, coincidentally $P_A/P_C = 1/6.5$ and $P_A/P_B = 1/3.7$. The estimated in-cloud NaCl deposition rate, D , is calculated from equation (1) and is found to be 69.3 kg/ha/yr.

Table 6 lists the volume of rainwater collected at site 1 and the rainfall amount as recorded by the Bureau of Meteorology rain gauge for the same

time interval. Also listed is the calibration ratio, r , of rainwater collected to rainfall measured. The relative constancy of this ratio over the 23 collections is reasonably good, considering that Macquarie Island experiences much wind, making precipitation collections difficult even at the best of times. Although only site 1 had a rain gauge, the calibration ratio is taken to be valid for all collection sites. The mean value of r was found to be 45.9 ml/mm, with a standard deviation of 7.8 ml/mm.

TABLE 4
Estimated mean rainfall rates for sites 1, 2 and 3

Site	Rainfall rate*	
	Mean percentage difference WRT site	Estimated mean R (mmhr ⁻¹)
1	—	0.402 (0.060)
2	25.4 (13.5)	0.504 (0.456)
3	42.0 (10.2)	0.571 (0.443)

* Numbers in parentheses give one standard deviation about the mean.

TABLE 5

Summary of deposition and rainfall rate, and mean particle concentration for various altitudes over Macquarie Island

Region	Total NaCl deposited at base of region	Estimated rainfall rate within region	Estimated particle concentration within region
	P (kg/ha/yr)	R (mmhr ⁻¹)	n (cm ⁻³)
A	370 (205)	0.571 (0.443)	200 (121)
B	655 (798)	0.504 (0.456)	376 (400)
C	2423 (2539)	0.402 (0.060)	1182 (1198)

* Numbers in parentheses indicate standard deviation of variable.

TABLE 6

Summary of rainwater and rainfall data for site 1

Date	Rainwater collected (m)	Rainfall recorded (mm)	Calibration ratio, r (ml/mm)
18 Dec 1985	683.5	15.2	45.0
18 Dec 1985	163.1	3.3	49.4
27 Dec 1985	92.6	2.1	44.1
29 Dec 1985	302.6	6.8	44.5
30 Dec 1985	300.2	6.2	48.4
31 Dec 1985	53.2	1.4	38.1
2 Jan 1986	326.3	6.0	51.0
3 Jan 1986	434.0	9.8	44.3
9 Jan 1986	167.2	6.2	27.0
13 Jan 1986	159.1	3.6	44.2
17 Jan 1986	898.0	15.6	57.6
1 Feb 1986	760.8	29.0	26.2
6 Feb 1986	409.6	8.6	47.6
11 Feb 1986	226.1	6.0	37.7
23 Feb 1986	3728.9	67.6	55.2
3 Mar 1986	2130.9	50.0	42.6
7 Mar 1986	846.1	16.6	51.0
9 Mar 1986	133.4	2.6	51.3
10 Mar 1986	255.5	5.0	51.1
25 Mar 1986	2359.4	51.0	46.3
29 Mar 1986	752.9	13.8	54.6
8 Apr 1986	1879.7	39.9	47.1
9 Apr 1986	220.0	4.3	51.2
Mean			45.9
Standard deviation			7.8

TABLE 7

NaCl concentrations collected from all available sites at approximately the same time

Collection date/period*	Collection site	[NaCl] (mm)	Percentage difference WRT site 1
29 Dec 1985	1	1520	-
(2)	2	853	-43.9
2 Jan 1986	1	5819	-
(3)	2	1279	-78.0
	3	1393	-76.1
9 Jan 1986	1	2964	-
(6)	2	588	-80.2
	3	696	-76.5
13 Jan 1986	1	1166	-
(4)	2	553	-52.6
	3	540	-53.7
23 Feb 1986	1	1606	-
(12)	2	743	-53.7
	4	319	-80.1

* Numbers in parentheses indicate sample period, in days, over which collections were made.

Note that converting from particle concentration in mg/l to kg/ha/yr simply requires multiplying concentration values by the conversion factor C, where

$$C = r(\text{ml/mm}) \times 10^{-3}(\text{l/ml}) \times \rho(\text{mm/yr}) \times 10^4(\text{m}^2/\text{ha}) \times 10^{-6}(\text{kg/mg}) \times \text{a}^{-1}(\text{m}^{-2})$$

where ρ is the yearly rainfall rate and a is the collection container area of 0.0511 m^2 . C needs to be determined for each site, since ρ differs.

TABLE 8

Ionic concentrations in rainwater from all collection sites

Collection date	[Cl ⁻]/[SO ₄ ²⁻]			
	Site 1	Site 2	Site 3	Site 4
18 Dec 85	17.1			
18 Dec 85	17.8			
19 Dec 85		18.5		
27 Dec 85	19.1	16.2		
29 Dec 85	19.2	18.3	18.2	
30 Dec 85	19.2	20.6	18.8	
31 Dec 85	19.0			
2 Jan 86	26.1	18.8	18.5	
3 Jan 86	18.4	14.3	12.5	
9 Jan 86	19.4	17.1	16.7	
13 Jan 86	18.0	16.1	16.4	
17 Jan 86	17.8			
18 Jan 86		16.8		
1 Feb 86	19.4			
2 Feb 86		18.4		
6 Feb 86	18.2			
7 Feb 86			17.0	
11 Feb 86	18.8	17.7		
13 Feb 86				17.8
23 Feb 86	19.1	17.7		15.7
3 Mar 86	18.9			
4 Mar 86		19.6		
7 Mar 86	19.6			
9 Mar 86	20.0			
10 Mar 86	20.4			
16 Mar 86		19.4		
25 Mar 86	19.9			
29 Mar 86	19.8	21.2		
3 Apr 86		19.0		
8 Apr 86	19.3			
9 Apr 86	19.4	19.4		

Table 7 lists mean NaCl concentrations for various sites of rainwater samples spanning concurrent periods of time. The collection date and site and percentage difference of [NaCl] with respect to site 1 are also given. Note that, on average, the percentage difference and standard deviations of [NaCl] between sites 1 and 2, and sites 1 and 3 are (61%, 16.5%) and (68.8%, 13.1%) respectively. Site 1 has consistently higher NaCl concentrations than sites 2 and 3. This is to be expected anyway, because the scavenging coefficient, A , decreases with increasing altitude since it is directly proportional to the vertical sea-salt concentration, as shown in table 5.

TABLE 9

NaCl concentration in individual water samples for all collection sites

Collection date	Equivalent NaCl deposited in rain (mg/l)			
	Site 1	Site 2	Site 3	Site 4
18 Dec 85	35.4			
	34.8			
19 Dec 85		40.4		
27 Dec 85	238.0	55.4		
29 Dec 85	88.8	49.8	36.2	
30 Dec 85	66.9	28.6	37.6	
31 Dec 85	645.8			
2 Jan 86	292.2	74.7	81.4	
3 Jan 86	27.9	6.2	6.5	
9 Jan 86	173.2	34.4	40.6	
13 Jan 86	68.1	32.3	31.6	
17 Jan 86	51.6			
18 Jan 86		25.5		
1 Feb 86	290.4			
2 Feb 86		118.6		
6 Feb 86	91.3			
7 Feb 86			50.9	
11 Feb 86	562.7	83.2		
13 Feb 86				72.1
23 Feb 86	93.8	43.4		18.6
3 Mar 86	50.2			
4 Mar 86		22.4		
7 Mar 86	455.0			
9 Mar 86	901.3			
10 Mar 86	123.8			
16 Mar 86		99.0		
25 Mar 86	595.0			
29 Mar 86	939.0	391.0		
3 Apr 86		33.4		
8 Apr 86	118.6			
9 Apr 86	185.0	85.6		

If n_x is now replaced by $n_x' = (n_A + n_B + n_C)/3$ and a recalculation of Q_M is made, then a more accurate theoretical estimate of Q_m can be made, based on a better estimate for n . n_x' is taken to be the mean NaCl particle concentration over site 1, averaged over regions A, B and C, a simple assumption which later proves to be a good estimate. Let Q_m' stand for this new theoretical estimate, which can be simply found from

$$Q_m' = Q_m(n_x'/n_x)$$

Q_m' and n_x' become equal to 1441 kg/ha/yr and 619 cm³ respectively. Q_m' is now the new theoretically derived value of NaCl deposition rate in

precipitation at site 1. It will be demonstrated in the next section that this compared favourably with actual measurements.

RESULTS

Unfortunately, due to an unknown and unspecified problem with the chemical analysis of rainwater, it was not possible to obtain consistent $[\text{Na}^+]$ values. However, $[\text{Cl}^-]$ and $[\text{SO}_4^{2-}]$ values were successfully determined. To demonstrate whether NaCl deposited in precipitation is marine derived, the ratio $[\text{Cl}^-]$ to $[\text{SO}_4^{2-}]$ was calculated and compared to the known ratio for seawater, a constant ratio of 19.3 (Millero 1974). Table 8 lists recovery dates, collection sites and the ratio $[\text{Cl}^-]$ to $[\text{SO}_4^{2-}]$ for all water samples used in the ensuing analysis. For sites 1, 2 and 3 the mean $[\text{Cl}^-]$ to $[\text{SO}_4^{2-}]$ ratio and standard deviation values are (19.3, 1.7), (18.2, 1.7) and (16.9, 2.1) respectively. Sulphates being of a higher molecular weight would be expected to have a different scavenging coefficient dependence with altitude. In fact, the trend appears to be that the ratio $[\text{Cl}^-]$ to $[\text{SO}_4^{2-}]$ decreases with increasing altitude. However, the result appears to be consistent with Evans' (1970), Jenkin's (1972) and Croome's (1984) conclusions, in that NaCl, at least, is marine derived in precipitation over Macquarie Island. Table 9 lists date, collection site and $[\text{NaCl}]$ for each sample collected. Much variability in concentration is evident and is

directly attributed to local weather conditions, such as humidity and wind speed. This variability is not examined in this study, as not enough is known about these conditions at sites 2, 3 and 4. Mean values of $[\text{NaCl}]$, as collected for each site, are listed in table 10 along with the equivalent deposition rate for NaCl.

It was calculated by Evans (1970) that 24 000 kg/ha/yr of NaCl are deposited in precipitation over site 1, and similarly 7999 and 565 kg/ha/yr of NaCl over site 2 and site 3 respectively, by Jenkin (1972). These amounts for sites 1 and 2 are in gross disagreement with those obtained in the present study. This discrepancy is either due to inaccurate chemical analysis or faulty collection procedures. For example, Evans does not describe the procedure used in decanting rainwater samples, nor if they were fixed against degradation. It is quite possible that either too few samples were collected and/or analysed or that too many rain events occurred before a collection was made. Averaging of $[\text{NaCl}]$ for too few rainwater samples may indeed have revealed such a high deposition rate. The variability in measured deposition rates in this study has been shown to be quite high, but it is not high enough to account for a tenfold variation in Evans' data for site 1. If a collection container is left unserviced for a long time it is quite possible that some evaporation of water can occur, due to wind and sun action; also dry salt particles can attach themselves to the collection container during non-precipitation

TABLE 10

Mean NaCl concentration from all samples for each collection site and their equivalent deposition rates, along with previously determined deposition rates by Evans (E) and Jenkin (J)*

Collection site	Number of rainwater collections	$[\text{NaCl}]$ (mg/l)	Calculated deposition rate of NaCl (kg/ha/yr)	
			Mallis	Evans and Jenkin
1	23	266.5 (279.5)	2423 (2539)	24 016 (E) [13]
2	17	72.0 (87.6)	654.5 (798)	7999 (J) [66]
3	7	40.7 (22.5)	370 (205)	565 (J) [50]
4	2	45.4 (-)	413 (-)	630 (E)

* Numbers in square brackets give the known number of continuous days over which rainwater was collected. Parentheses contain the standard deviation of the mean. No standard deviations are given for site 4, because only two samples were collected, and so no statistical significance could be claimed.

periods. Either possibility, or a combination, can lead to unrealistically high levels of NaCl; hence, in the present study, the collection of rainwater samples every few days from all sites. Thus, it is claimed that data collected by the present study and listed in table 10 are much more representative of actual deposition rates than Evans' and some of Jenkin's data. Evans' data are also questionable on the grounds that Millero (1974) gives average world seawater [NaCl] as 30 120 mg/l. This compares with 2674 mg/l from Evans' data for site 1, which is some 9% of that of seawater. This figure seems quite large and unrealistic.

CONCLUSION

As a result of the present studies, it is claimed that deposition of scavenged airborne NaCl particles over Macquarie Island in precipitation is almost entirely due to dry sea salt and not sea spray. Although it cannot be explicitly proven that this finding is valid, it is encouraging that the theoretically predicted deposition rate of 1441 kg/ha/yr falling on site 1 is 60% of that actually recorded, namely 2423 kg/ha/yr. Considering that the observed standard deviation was 2539 kg/ha/yr, the theoretically derived value seems an acceptable figure. It appears that Evans' (1970) and Jenkin's (1972) data previously contributed to false inferences regarding NaCl scavenging. It is now concluded that scavenging of airborne (dry) NaCl particles can alone successfully account for the total NaCl deposited due to in- and below-cloud scavenging over Macquarie Island, and there is no need to suppose that scavenging of sea spray is responsible for any major component of the total salt budget. Experimental data, collected to test previous theoretical calculations made of this problem, seem to verify the above conclusion.

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