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## IDENTIFICATION OF COMPONENT SOURCES IN ANTARCTIC SNOW BY FACTOR ANALYSIS

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The concentration values of twelve components determined in snow samples collected in two snow-pits around Terra Nova Bay, Antarctica, are Factor Analysed. The factors are interpreted as different sources affecting the area.

The marine aerosol factor dominates the data set structure, but two other factors can be clearly identified for both stations, one reflecting seasonal and the other linked to biogenic sulfur cycle. Distinct features characterising the two stations emerge from the factor analysis applied to a reduced data set, in which a limited number of samples with a very high sea-salt loading are excluded

*Keywords:* Snow composition; Antarctica; factor analysis; marine contribution

### INTRODUCTION

The polar areas have been identified as ideal sites for the study of the global scale transport of pollutants emitted mainly in the highly populated and industrialised countries of the northern hemisphere.

Antarctica in particular, due to its isolation from other continental masses, is a laboratory of interest for the study of the bio- and geo-chemical cycles of substances of natural origin, such as the marine and crustal aerosol, and the transport of pollutants on the global scale<sup>[1-3]</sup>.

The systematic collection of samples and meteorological data is unfortunately possible only near the very few permanent stations and limited usually to the sunlit periods due to logistic impediments, but the collection of ice cores and surface snow-pit samples provides records covering complete annual cycles dating

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back to tens of years for snow-pits and hundreds of thousand of years for ice-cores.

The chemical information obtained from the analysis of the different layers can be used to reconstruct the stratification sequence and to follow the temporal evolution of the contributions of the different aerosol sources to the precipitation composition.

The aim of the present paper is to identify the common factors of variance of two data sets relative to the analytical concentrations determined in samples collected in two snow-pits around the Italian base of Terra Nova Bay to identify the factors as different aerosol sources or meteorological parameters influencing the area, and to point out differences in the data set structure that can be ascribed to the distinct geographical position of the sampled sites.

The source resolution in the data set is achieved by Principal Factor Analysis (PFA), a multivariate statistical technique which is widely used to solve similar problems.

## EXPERIMENTAL

During the 1990–91 Antarctica field season, as part of the Italian Program of Antarctic Research (PNRA), two snow-pits, about 2 m deep, were excavated in two sites, Styx Glacier Plateau (73°51'56"S, 163°41'33"E) and McCarthy Ridge (74°32'57"S, 162°56'29"E), in the Northern Victoria Land, Antarctica (Figure 1). The two stations were chosen at different altitudes to assess the influence of the elevation on the precipitation composition. The samples were collected inserting pre-cleaned polyethylene vials in the vertical walls of the pit: two series of samples were collected for each station, using 16 and 35 mm internal diameter vials. The vials were sealed in polyethylene bags and shipped to the laboratory in Florence, where they were allowed to melt at room temperature in a laminar flow hood class 100 and analysed by Ion Chromatography and Flow Injection Analysis. Strictly anti contamination procedures were followed throughout the sampling and analytical procedures<sup>[4]</sup>.

The principal (chlorides, nitrates and sulphates) and trace (bromides, phosphates, fluorides, formates and methansulphonates) anions together with hydrogen peroxide, which is the seasonal tracer used to reconstruct the annual layers, were determined in both series of vials, whereas the cations sodium, potassium, magnesium and calcium were determined only for the 35 mm vials.

The characteristics of the instrumentation and the analytical method parameters (reproducibility and detection limit) are reported elsewhere<sup>[5]</sup> as well as the results of the analysis of the 16 mm vials<sup>[6]</sup>. Here we focus the discussion to the

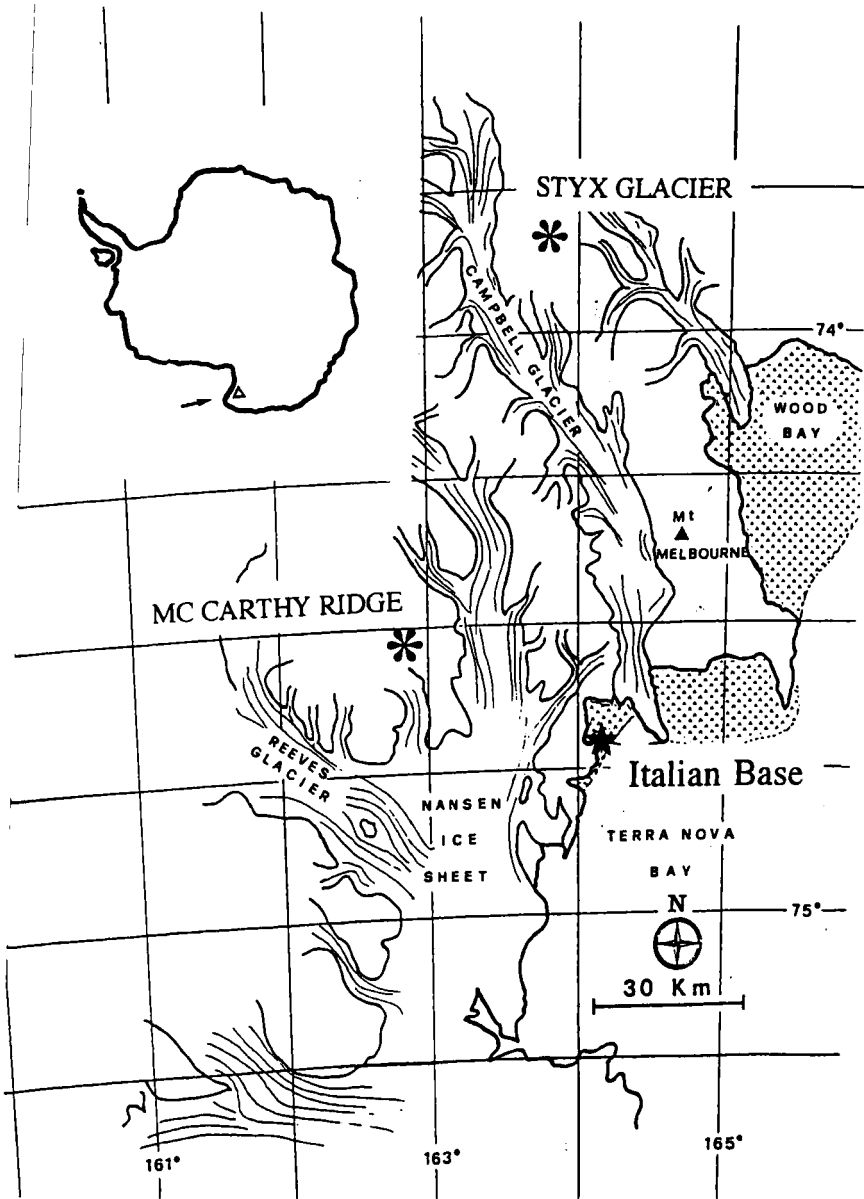


FIGURE 1 Map of a part of Northern Victoria Land showing the position of Terra Nova Bay base and of the two sampling stations

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35 mm vials data set, far more complete and thus better suited for a statistical evaluation. The vertical profiles obtained for the ions determined in both series were compared and the almost perfect match between the corresponding time series assures the analysis' reliability. Table I and II show the summary statistics for the samples collected at Styx Glacier Pl. and McCarthy Ridge. The variables bromides and phosphates were excluded from the PFA due the high percentage of values below the detection limit as reported in the tables. The data sets used for the PFA are thus composed: 46 samples for Styx Glacier Pl., relative to depositions accumulated during 1986–90 and 58 samples for McCarthy Ridge representing depositions of the period 1987–90.

TABLE I Summary statistics of measured ion concentration  $\mu\text{g/Kg}$  for Styx Glacier Plateau

<i>Variable</i>	<i>Mean</i>	<i>Med</i>	<i>SE</i>	<i>Min</i>	<i>Max</i>	<i>Sk</i>	<i>Ku</i>	<i>Samples below D.L.</i>
$\text{Cl}^-$	918	232	283	70	9274	3.2	9.9	0
$\text{NO}_3^-$	78.1	58	8.77	17	310	2.1	4.74	0
$\text{Br}^-$	1.18	1	0.16	<D.L.	3.2	1.17	0.89	49
$\text{H}_2\text{PO}_4^-$	9	5.4	1.92	<D.L.	49	2.7	6.72	28
$\text{SO}_4^{2-}$	173	74	41.9	26	1390	3.17	9.78	0
$\text{nssSO}_4^{2-*}$	33.6	28.3	7.46	-80.2	128.8	-0.18	0.13	0
$\text{F}^-$	1.12	0.76	0.16	0.12	6.23	3.01	11.36	0
$\text{HCOO}^-$	4.01	3.65	0.21	1.9	8.8	1.35	2.33	0
$\text{CH}_3\text{SO}_3^-$	11.6	6.2	1.94	1.3	58.5	2.29	5.31	0
$\text{Na}^+$	555	126	176	21	5801	3.15	9.64	0
$\text{K}^+$	34.3	15.6	6.95	4.3	216	2.38	5.26	0
$\text{Mg}^{2+}$	62.4	15.2	18.9	3.3	588	3.01	8.79	0
$\text{Ca}^{2+}$	64.5	48.7	7.8	24	256	2.35	5.44	0
$\text{H}_2\text{O}_2$	42.6	32.1	6.71	0.1	181	1.29	1.48	0

Med = median; SE = standard error of mean; D.L. = detection limit. Sk and Ku = skewness and kurtosis of data distributions.

\* = nssSO<sub>4</sub> is non sea salt sulphate or sulphate in excess of sea water based on the sodium content.

## RESULTS AND DISCUSSION

The multivariate statistical technique of Principal Components or Factor Analysis has been often used to resolve the sources contributing to the measured

concentrations of constituents in aerosol samples collected in urban and rural areas<sup>[7-11]</sup>.

TABLE II Summary statistics of measured ion concentration  $\mu\text{g/Kg}$  for McCarthy Ridge

<i>Variable</i>	<i>Mean</i>	<i>Med</i>	<i>SE</i>	<i>Min</i>	<i>Max</i>	<i>Sk</i>	<i>Ku</i>	<i>Samples below D.L.</i>
$\text{Cl}^-$	1668	675	402	61	19763	4.22	21.65	0
$\text{NO}_3^-$	51.9	41	5.48	14	234	2.38	6.61	0
$\text{Br}^-$	3.05	1.3	1.09	<D.L.	49.4	6.1	39.27	22
$\text{H}_2\text{PO}_4^-$	5.69	4.35	0.79	<D.L.	33.8	3.68	17.18	21
$\text{SO}_4^{2-}$	328	213	52.16	48	2618	3.97	19.68	0
$\text{nssSO}_4^{2-*}$	98	74.1	12.77	-80.9	347.3	0.48	-0.4	0
$\text{F}^-$	0.851	0.62	0.09	0.03	2.6	1.05	-0.01	0
$\text{HCOO}^-$	4.39	4.05	0.26	1.5	11.5	1.26	2.03	0
$\text{CH}_3\text{SO}_3^-$	38.6	26.2	4.86	1.7	130.1	1.11	0.24	0
$\text{Na}^+$	914.7	409	217	46	10747	4.27	22.26	0
$\text{K}^+$	61.3	46.6	8.03	6.7	421.4	3.98	21.13	0
$\text{Mg}^{2+}$	104	45.7	24.9	7.1	1260	4.48	24.54	0
$\text{Ca}^{2+}$	74.9	55.8	8.95	21.5	464.1	3.78	18.54	0
$\text{H}_2\text{O}_2$	27.4	14.55	4.44	0.5	143.9	1.64	2.54	0

Med = median; SE = standard error of mean; D.L. = detection limit. Sk and Ku = skewness and kurtosis of data distributions.

\* = nssSO<sub>4</sub> is non sea salt sulphate or sulphate in excess of sea water based on the sodium content.

This technique has been successfully applied to resolve the sources of aerosol samples collected in the Arctic Region<sup>[12-13]</sup> especially when applied to ionic rather than elemental concentrations, as in this case. The associations of particular variables in the components can be interpreted as the result of chemical interactions between the ions in the atmosphere at Barrow (Alaska)<sup>[14]</sup> and besides the sulphur can be apportioned between sulphate and methansulphonate (MSA) thus aiding the identification of the biogenic marine source of sulphur.

Boutron and Martin<sup>[15]</sup> applied the technique to the metal concentrations determined in snow and ice samples collected at different sites in Antarctica, both on the coast and the continent, to investigate the different contributions of marine and crustal aerosol to the concentrations of alkaline and alkaline-earth metals as a function of distance from the coast.

The PFA technique can be applied to a set of variables to reduce the dimensionality of the problem, substituting a set of correlated base vectors, with an orthogonal set, the Principal Factors, which are a linear combination of the original variables, chosen in such a way that the first factor accounts for as large a

percent of the original variance as possible, while the subsequent factors explain as much as possible of the residual variance with the constraint of being orthogonal to the factors already extracted.

If  $k = 1, 2, \dots, m$  is the number of variables considering the eigenvalue defined as:

$$\lambda_j = \sum_{k=1}^m a_{kj}^2 \quad \text{where } j = 1, 2, \dots, p, \dots, m$$

and the communality,  $h_k^2$ , as the sum

$$h_k^2 = \sum_{j=1}^p a_{kj}^2 \quad \text{where } p \leq m \text{ is the number of significant factors}$$

in Tables III and IV the eigenvalues and the relative percent of variance are shown and explained by each factor, together with the percent cumulative variance for the two stations.

TABLE III Factor analysis results for the complete data set at Styx Glacier

<i>Factor</i>	<i>Eigen value</i>	<i>% Var</i>	<i>% Cum Var</i>
1	6.82632	56.9	56.9
2	2.58656	21.6	78.4
3	1.08026	9	87.4
4	0.73418	6.1	93.6
5	0.38173	3.2	96.7
6	0.20529	1.7	98.5
7	0.10240	0.9	99.3
8	0.05161	0.4	99.7
9	0.02777	0.2	100
10	0.00286	0	100
11	0.00104	0	100
12	-0.00002	0	100

The tables report also the initial estimate of communality for each variable, SMC = Squared Multiple correlation.

The problem with the Factor Analysis is to find the number of significant factors. In this case the number was chosen so that the resulting model is sufficiently representative while avoiding redundancy at the same time.

TABLE IV Factor analysis results for the complete data set at McCarthy Ridge

Factor	Eigen value	% Var	% Cum Var
1	6.80956	56.7	56.7
2	2.10443	17.5	74.3
3	1.16851	9.7	84
4	0.90293	7.5	91.5
5	0.46513	3.9	95.4
6	0.35701	3	98.4
7	0.08658	0.7	99.1
8	0.06695	0.6	99.7
9	0.03600	0.3	100
10	0.00188	0	100
11	0.00101	0	100
12	0.00000	0	100

Four factors were found significant for both stations and Tables V and VI report the varimax rotated loadings for the two data sets.

TABLE V Varimax rotated loading matrix at Styx Glacier

Variable	PF1	PF2	PF3	PF4	Communality
Cl <sup>-</sup>	0.9756	-0.07198	0.14312	-0.03043	0.97839
NO <sub>3</sub> <sup>-</sup>	0.21114	0.89665	0.24476	0.03763	0.90989
SO <sub>4</sub> <sup>2-</sup>	0.97404	0.02704	0.16911	0.04997	0.98058
nssSO <sub>4</sub> <sup>2-*</sup>	-0.34466	0.72678	0.18178	0.4908	0.92092
F <sup>-</sup>	0.37503	0.04692	0.87403	0.00129	0.90678
HCOO <sup>-</sup>	0.78398	-0.19549	-0.30354	0.1268	0.76106
CH <sub>3</sub> SO <sub>3</sub> <sup>-</sup>	0.10904	0.228	-0.03384	0.95739	0.98161
Na <sup>+</sup>	0.97914	-0.09485	0.12998	-0.03412	0.98577
K <sup>+</sup>	0.92358	-0.08586	0.2978	-0.0443	0.95102
Mg <sup>2+</sup>	0.96639	-0.0384	0.22528	-0.01413	0.98633
Ca <sup>2+</sup>	0.91792	0.15534	0.31297	0.04152	0.96638
H <sub>2</sub> O <sub>2</sub>	-0.18807	0.84777	-0.34319	0.16351	0.89859
	marine aerosol	seasonal	fluoride	biogenic sulphur	



TABLE VI Varimax rotated loading matrix at McCarthy Ridge

Variable	PF1	PF2	PF3	PF4	Communality
Cl <sup>-</sup>	0.96377	-0.12418	0.20869	-0.06367	0.99187867
NO <sub>3</sub> <sup>-</sup>	-0.00158	0.05817	0.05186	0.68783	0.47918581
SO <sub>4</sub> <sup>2-</sup>	0.98727	0.03849	0.11948	0.08642	0.99792742
nssSO <sub>4</sub> <sup>2-*</sup>	-0.09119	0.70562	-0.40542	0.58275	1.01017814
F <sup>-</sup>	0.44173	-0.16849	0.64999	0.16248	0.67240102
HCOO <sup>-</sup>	0.41209	-0.56364	-0.02082	0.10902	0.49982705
CH <sub>3</sub> SO <sub>3</sub> <sup>-</sup>	-0.02527	0.73601	-0.51458	0.25404	0.87167819
Na <sup>+</sup>	0.96649	-0.12853	0.20938	-0.05385	0.99736269
K <sup>+</sup>	0.91134	-0.25436	0.14399	-0.00879	0.91604999
Mg <sup>2+</sup>	0.96694	-0.13657	0.19292	-0.0624	0.99473621
Ca <sup>2+</sup>	0.92608	-0.20953	0.25478	0.05431	0.96938941
H <sub>2</sub> O <sub>2</sub>	-0.16753	0.11195	-0.62217	0.02935	0.42855603
	marine aerosol	biogenic sulphur	seasonal	nitrate	

The first factor in both cases is the marine aerosol source, as identified by the loadings of the variables chlorides, magnesium and sodium; the variables sulphates, potassium and calcium are also strongly loaded on this component, which explains a great percentage of the variance for these variables.

The second factor for Styx Glacier Plateau and the third for McCarthy Ridge are identified as seasonal factors. For Styx Glacier Plateau this factor is mainly loaded with nitrates, H<sub>2</sub>O<sub>2</sub> and nss-sulphates; the first two compounds derive from photochemical reactions in the atmosphere whereas the third has a significant biogenic source so that they all show seasonal cycles coherent with the winter-summer sequence in Antarctica<sup>[16-17]</sup>. For McCarthy Ridge the seasonal factor is positively correlated with F<sup>-</sup> and inversely with H<sub>2</sub>O<sub>2</sub>. The fluorides source is unidentified but its temporal profile shows a seasonal signal with winter maxima and this explains its inverse relationship with H<sub>2</sub>O<sub>2</sub>.

The second factor for McCarthy Ridge and the fourth for Styx Glacier Plateau is mainly related with MSA and nssulphate and is interpreted as a biogenic sulphur source. The structure of the two data sets is different for the variables nitrate, that is an independent factor for McCarthy Ridge (the fourth factor) and for fluorides and formates, whose associations with both the marine as well as other factors are difficult to interpret. Both stations are mainly affected by the

marine aerosol source, strongly correlated with chlorides, magnesium, sodium, potassium, sulphates and calcium. However, the ratios of the concentrations of these variables to sodium, assumed to derive solely from sea-spray, are distributed around the standard sea water value for magnesium and chlorides as reported in Table VII, while for potassium, calcium and sulphates the ratios are close to the theoretical value only for a limited number of samples with high sodium loading, and are decidedly higher for the remaining samples, as can be seen from the average values reported in the table.

TABLE VII Distribution parameters of the ratios between concentrations of reported variables and their corresponding value measured in standard sea-water

<i>Styx Glacier</i>	<i>Mean</i>	<i>Median</i>	<i>Stand. Dev</i>	<i>Sea-water value (a)</i>
Cl/Na	2.2	1.9	0.817	1.80
Mg/Na	0.13	0.12	0.057	0.12
SO <sub>4</sub> /Na	0.84	0.55	0.798	0.252
K/Na	0.17	0.090	0.179	0.037
Ca/Na	0.52	0.34	0.590	0.038
<i>McCarthy Ridge</i>	<i>Mean</i>	<i>Median</i>	<i>Stand. Dev</i>	
Cl/Na	1.8	1.8	0.309	
Mg/Na	0.12	0.11	0.021	
SO <sub>4</sub> /Na	0.72	0.46	0.739	
K/Na	0.13	0.11	0.090	
Ca/Na	0.17	0.14	0.108	

(a) T.R.S. Wilson, in *Chemical Oceanography*, Riley and Skirrow eds., Academic Press, 1975.

This suggests that while chlorides and magnesium are derived almost completely from sea-spray, other sources may be influencing the measured concentrations of potassium, calcium and sulphates, and these contributions become evident when the sea-spray content is minimum. The samples are divided in two groups, one with high and the other, more numerous, with low sodium content, according to the results of a Cluster Analysis applied to the Euclidean distances between the samples concentrations over the space defined by the variables sodium, chlorides, magnesium, potassium, calcium and sulphates. The resulting clusters, indicated by different markers, are plotted in Figure 2. The samples belonging to the first clusters, 35 and 48 samples respectively for Styx Glacier Plateau and McCarthy Ridge, are then subjected to a Factor analysis to try to identify these extra sources. The resulting varimax rotated loadings are shown in Tables VIII and IX.

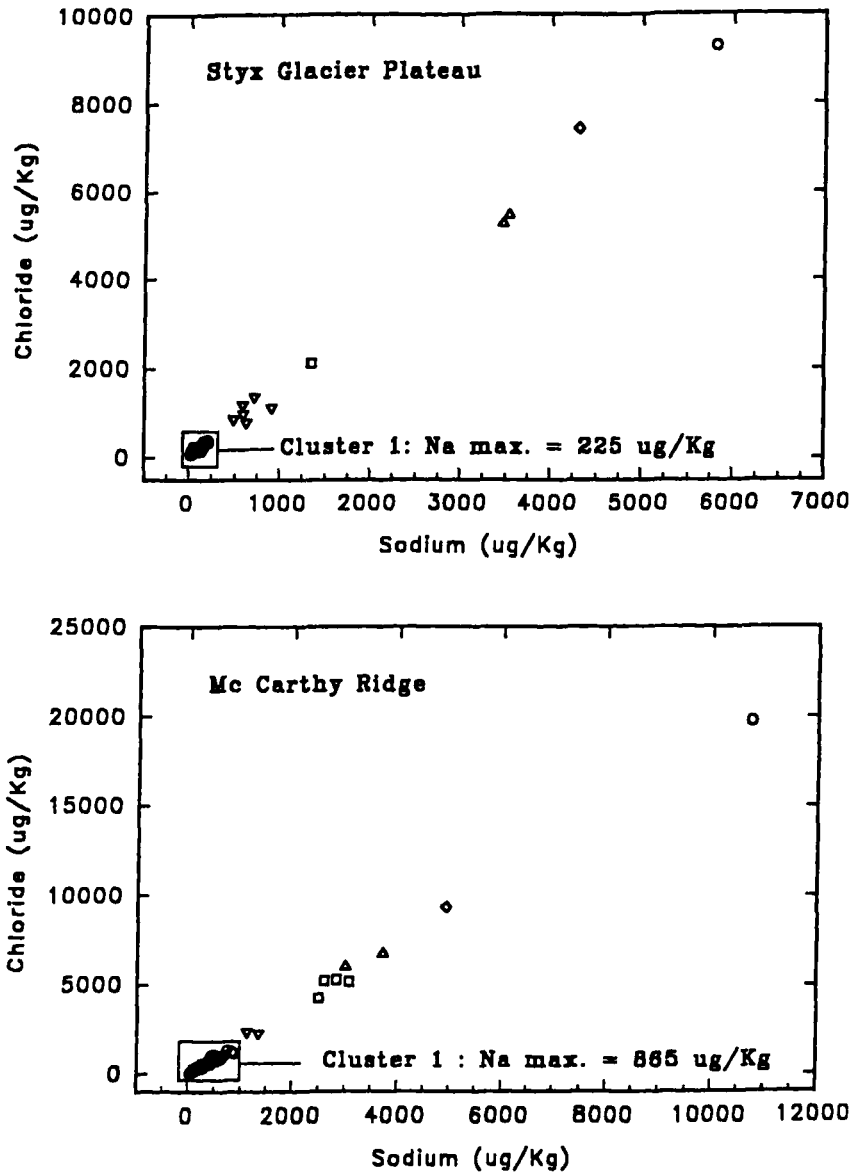


FIGURE 2 Samples' grouping resulting from the cluster analysis of the Euclidean distances of Na and Cl for Styx Glacier plateau and McCarthy Ridge

TABLE VIII Varimax rotated loading matrix for the reduced data set at Styx Glacier

<i>Variable</i>	<i>PF1</i>	<i>PF2</i>	<i>PF3</i>	<i>PF4</i>	<i>Communality</i>
Cl <sup>-</sup>	-0.05275	0.94744	-0.04986	0.12471	0.918464
NO <sub>3</sub> <sup>-</sup>	0.87684	0.05443	-0.15936	-0.01806	0.797533
SO <sub>4</sub> <sup>2-</sup>	0.85924	0.19341	0.39460	0.06233	0.935295
nssSO <sub>4</sub> <sup>2-*</sup>	0.89494	-0.22240	0.31335	0.03816	0.950024
F <sup>-</sup>	0.08033	0.52400	-0.28243	0.49014	0.601033
HCOO <sup>-</sup>	-0.03575	0.19548	0.77850	-0.01373	0.645741
CH <sub>3</sub> SO <sub>3</sub> <sup>-</sup>	0.47753	-0.10187	0.72285	0.08092	0.767473
Na <sup>+</sup>	-0.14169	0.94815	0.16070	0.05169	0.947561
K <sup>+</sup>	-0.17297	0.02409	0.10830	0.89516	0.843539
Mg <sup>2+</sup>	0.11709	0.93752	0.16777	-0.09154	0.929180
Ca <sup>2+</sup>	0.71978	0.16455	0.08196	-0.22482	0.602421
H <sub>2</sub> O <sub>2</sub>	0.82777	-0.26638	-0.05028	-0.12874	0.775264
	Seasonal	Marine aerosol	?	Potassium	

TABLE IX Varimax rotated loading matrix for the reduced data set at Mc Carthy Ridge

<i>Variable</i>	<i>PF1</i>	<i>PF2</i>	<i>PF3</i>	<i>PF4</i>	<i>Communality</i>
Cl <sup>-</sup>	0.95451	-0.02687	0.03504	0.08514	0.920288
NO <sub>3</sub> <sup>-</sup>	-0.29883	0.51942	0.18627	0.53892	0.684228
SO <sub>4</sub> <sup>2-</sup>	0.32536	0.93440	-0.07876	-0.01232	0.985317
nssSO <sub>4</sub> <sup>2-*</sup>	-0.17710	0.95865	-0.12620	-0.07187	0.971466
F <sup>-</sup>	0.14973	-0.14513	0.09731	0.83246	0.745941
HCOO <sup>-</sup>	-0.18181	-0.21799	0.84514	0.03550	0.796096
CH <sub>3</sub> SO <sub>3</sub> <sup>-</sup>	-0.06478	0.88390	-0.19483	-0.29229	0.908868
Na <sup>+</sup>	0.96904	0.02918	0.08221	0.11047	0.958852
K <sup>+</sup>	0.40036	-0.05291	0.73111	0.06756	0.702174
Mg <sup>2+</sup>	0.96626	-0.02758	-0.00019	0.08798	0.942160
Ca <sup>2+</sup>	0.66043	-0.00199	0.52568	0.31503	0.811755
H <sub>2</sub> O <sub>2</sub>	-0.43065	0.24828	0.03443	-0.64732	0.667311
	Marine aerosol	Biogenic sulphur	?	Seasonal	

The difference between the two stations is now evident. The first factor for Mc Carthy Ridge is still the marine aerosol component, which is now correlated only with sodium, chlorides and magnesium. This factor now becomes the second for Styx Glacier Plateau, in agreement with the fact that a higher elevation for this station will result in the condensation and precipitation of a great proportion of the sea-spray aerosol particles due to the adiabatic cooling of the oceanic air masses during the upslope transport from the sea level to the more elevated sites in Antarctica.

The total sulphates, that were loaded on the factor identified as sea spray in the analysis of complete data set, are now highly loaded in biogenic sulphur factor, together with MSA and nsssulphate. This suggests that the principal source of the sulphates for the samples with a limited sea spray contribution is biogenic. For Styx Glacier Plateau sulphates are associated with the seasonal factor, now loaded with nitrates, H<sub>2</sub>O<sub>2</sub>, nsssulphate and calcium, with a less contribution from MSA.

Based on these data sets it is not possible to determine if the association between calcium and nsssulphates is to be ascribed to a specific source or is the result of interactions between aerosol particles during transport. Crystals of calcium sulphate have been identified in aerosol samples collected in remote marine areas with the electron microprobe analysis technique<sup>[18–19]</sup> and they are supposed to be the result of the reactions between sulphuric acid and calcium carbonate particles emitted from the ocean surface into the atmosphere.

## CONCLUSIONS

The principal factor analysis applied to the two complete data sets shows that both stations are mainly influenced by the marine aerosol component. A detailed analysis of the distributions of the ratios between the variables heavily loaded on this component with sodium, however, suggests that this simplified picture is the result of the high influence of a limited number of samples with a high salt content on the correlation matrices, which is the basis of the Factor Analysis technique. Only when this particular group of sample is excluded the factor analysis model result in a more definite picture of the two data sets. This should be expected on theoretical grounds considering the influence that a different elevation has on the transport and depositional patterns of the aerosol sources affecting the area.

### **Acknowledgements**

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