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1 **Long-term monitoring of atmospheric pollution in Maritime Antarctic with lichen**  
2 ***Usnea aurantiaco-atra* (Jacq.) Bory: a magnetic and elemental study.**

3

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20

21 **Abstract**

22 Antarctica is a natural research laboratory thanks to its unique climate, geography, flora,  
23 and fauna. The conservation of Antarctica's environment is monitored through the  
24 Madrid Protocol; however, there are local pollution problems associated to human  
25 activities like research and tourism; in particular, there are negative impacts on air  
26 quality from the use of fossil fuels. In this work, we studied for the first time magnetic  
27 and elemental characteristics of lichen *Usnea aurantiaco-atra* (Jacq.) Bory collected in  
28 different years and from various sites in King George Island, Antarctic Peninsula, as  
29 well as some samples of its supporting substrate, for long-term monitoring of the  
30 atmospheric pollution. Several anthropogenic elements (Ni, Pb, Mo, Cd, and Zn) have  
31 been identified on sites close to human activities but also on sites far from it. We found  
32 that magnetic proxies from *U. aurantiaco-atra* samples show a spatial correlation with  
33 human influence (scientific bases or airstrips). We observed a correlation between  
34 magnetic parameters and Ni, and to a lesser extent with Cr, Co, V, and Ag. Results  
35 suggest that using these magnetic and elemental techniques, it is possible to implement  
36 monitoring with lichen *U. aurantiaco-atra* as a bioindicator for some elements of  
37 anthropogenic origin.

38

39

40 Key words: environmental magnetism, biomonitoring, anthropogenic particulate matter,  
41 elemental composition, Fildes Peninsula, King George Island.

42

## 43 **Introduction**

44 The Antarctica continent covers an area of 14062500 km<sup>2</sup> of which only 0.32% of its  
45 total area is free of ice. Vegetation grows on a constrained basis in coastal sectors free  
46 from ice and snow during the summer and in some rock outcrops located inland.  
47 Despite its remote situation and extreme conditions, human presence has existed in  
48 Antarctica for more than 200 years associated to whaling activity (Tin et al. 2009) and  
49 for more than 70 years associated to science and tourism. Nowadays, the human  
50 presence is represented by 112 research bases throughout the continent, which  
51 accommodate over 5000 people during the summer. In winter, 40 stations are in  
52 operation, which could accommodate 3916 people, who need water, fossil fuel  
53 consumption and management of wastes. Moreover, tourism has been continuously  
54 increasing; this caused the visit of over 50000 people during the season 2017-2018, with  
55 obvious environmental implications.

56 King George Island, Antarctic Peninsula, Maritime Antarctica stands out as “hot spot”  
57 receiving increasing pressure from human activities. Fildes Peninsula with about 29 km<sup>2</sup>  
58 of surface has a high biodiversity and is a major logistic centre for the northern Antarctic  
59 Peninsula; it supports the largest number of scientific bases (5 permanent from Chile,  
60 China, Russia, Uruguay) and shelters and an airfield for aircrafts arriving from the city  
61 of Punta Arenas, Chile.

62 There is no monitoring of air quality and certainly no inventory of sources in the Fildes  
63 Peninsula (Tin et al. 2009). Studies by Mishra et al. (2004) on total atmospheric aerosols  
64 (TSP, suspended particles smaller than 45-50 µm) from King Sejong base (Barton  
65 Peninsula) indicate that presence of the elements Bi, Cd, Co, Cr, Cu, Ni, V and Zn is

66 caused by anthropogenic activities associated with local sources. For Fildes peninsula,  
67 information is available on fractionated aerosols smaller than 3  $\mu\text{m}$ . The elements Na,  
68 Mg, Sr, Mn, Fe, Cr, Co and Ca have a natural origin and Pb, Ni, Zn, Cu and Cd, on the  
69 contrary, have an anthropogenic origin. The elements Pb, Cr, Sb, Cu and Mn are  
70 partially of extra continental origin. Ca, Ba, Ni and partially Zn and Cu are generated by  
71 the human activities in the island (Préndez et al. 2009, Carrasco & Préndez, 1991).

72 Mosses and lichens constitute the main biotic components of terrestrial ecosystems in  
73 Antarctica. Exposed biological surfaces, e.g., lichens, mosses and leaves, accumulate  
74 atmospheric particles, and are able to provide a record of location-specific and time-  
75 integrated information on local air quality (Bajpai et al., 2018). The relatively high  
76 humidity and mild temperatures found in maritime Antarctica, together with the absence  
77 of competition from higher plants, create the perfect place for stress-tolerant organisms  
78 such as lichens (Sancho & Pintado 2011). Lichens are good bio-accumulators of  
79 chemical elements transported by air (Préndez et al. 2006, Sancho et al. 2007) and can  
80 be used as bio-monitors of the atmospheric deposition of inorganic pollutant also in  
81 Antarctica (Poblet et al.1997). Nutrients and/or material derived from the humid and dry  
82 atmospheric precipitation deposit can accumulate on or around the thallus. The  
83 incorporation of elements through the thallus surface depends on many factors such as  
84 the nature of the element, morphologic characteristics of the surface, and environmental  
85 parameters (Wolterbeek 2002). When using lichens as bioindicators, it is necessary also  
86 to evaluate the contribution of the substrate in which they grow in order to determine  
87 only the atmospheric contribution in nutrition and accumulation of contaminants in the  
88 thallus. Poblet et al. (1997) have shown that *Usnea aurantiaco-atra* (Jacq.) Bory

89 (currently *Neuropogon aurantiaco-ater*) is a convenient organism for developing a bio-  
90 monitoring of metals in atmospheric aerosols in the area of the South Shetland Islands.  
91 The United States Forest Service (2010) provides an extensive database of air quality in  
92 relation to biomonitoring through lichens with tables including threshold levels of  
93 chemical elements present in Oregon, Washington and in Alaska's national forests. In  
94 our knowledge, there is no similar database for Antarctica.

95 Atmospheric pollution, in particular urban particulate matter (PM), often contains levels  
96 of magnetic minerals, e.g., iron oxides like magnetite, hematite and maghemite (e.g.,  
97 Matzka & Maher, 1999) that are easily measurable magnetically. This PM can  
98 accumulate on exposed biological surfaces and deliver accumulated information on local  
99 air quality. A group of trace metals, such as Zn, Cd, Pb, Ni, Cu and Cr, are often directly  
100 associated with magnetic PM, due to their incorporation in the mineral structure during  
101 combustion processes (Cao et al. 2015). Therefore, the magnetic signal may act not only  
102 as a PM proxy but be of direct, often health-related, interest in itself.

103 Only a few magnetic measurements have been carried out on lichens, in situ or as  
104 transplants (see Hofman et al. 2017 for a review). These studies show that magnetic  
105 properties are good proxies for anthropogenic atmospheric pollution, though they are  
106 species-dependent.

107 The aim of this study was to evaluate the use of *U. aurantiaco-atera* lichen as  
108 bioindicator of air quality in the Fildes Peninsula over a period of 13 years (1997-2010)  
109 using magnetic and elemental techniques.

110

111 **Sampling**

112 *Substrates and Lichens*

113 Five-g samples of *U. aurantiaco-atra* were carefully collected in some summer/fall  
114 seasons between 1997 and 2010. Samples were taken from 1\*1 m<sup>2</sup> demarcated and  
115 protected sites (Table I), then air-dried in Antarctica, stored in clean paper bags,  
116 transported to the laboratory for analyses, and kept isolated from external influences.  
117 Lichen thalli, when dried, do not decompose. In the laboratory, duplicates of all lichen  
118 samples were sorted, using only the thinnest thalli, and dead and senescent tissue,  
119 superficial dust, soil particles, and small stones were manually removed using a Teflon  
120 clamp. Samples were dried at 90°C for 24 h, mashed in an agate mortar, and stored in  
121 sterile Petri dishes to provide samples to the different analysis.

122 In parallel, 2 or 3 samples of the substrate (first 5 cm of the substrate) were taken  
123 together with the lichens. Composite substrate samples were assembled and were air-  
124 dried in Antarctica and packed in paper bags further stored in plastic containers at -18  
125 C°.

126 We lack samples of substrate or lichens for some years and/or sites, not because the  
127 samples were not taken, but because they were used in other analyzes over the years and  
128 there was no more left.

129 *Sampling sites*

130 King George Island is located between 61°50' and 62°15'S and 57°30 and 59°01'W; it  
131 belongs to the South Shetlands Islands and is located near the northern part of the  
132 Antarctic Peninsula (Fig. 1). The Collins Glacier covers practically the entire island. The  
133 rest of the surface, mostly along the coast, is composed of volcanic rocks (basalts and/or

134 andesites) which form cliffs about 100 m high. The substrate is composed of material  
135 with coarse granulometry.

136 The sampling points are localized in sites close to potential human emission sources,  
137 such as the aerodrome and scientific bases, and more isolated sites where there should be  
138 less human influence, such as the Ardley peninsula and the Collins Glacier (Fig. 1).

139 The sites that are close to emission sources are: Site Aerodromo, located near the  
140 Aerodrome Teniente Rodolfo Marsh, which is used by many the scientific bases on the  
141 island; Site LARC, located next to the Antarctic Laboratory of Cosmic Radiation,  
142 University of Chile that is installed on top of a hill of about 40 m above sea level (a.s.l),  
143 close to two Chilean bases; Site Base China which is located about 100 m southeast of  
144 the Great Wall Chinese base station, under construction/extension all along the sampling  
145 period; Site Aerodromo MET, located in the vicinity at east side of the Aerodrome T.R.  
146 Marsh.

147 The sites chosen far away from possible anthropogenic emissions are: Site Ardley,  
148 located on a small peninsula located southeast of Fildes and connected by a sand dam,  
149 which is a nesting site for a wide variety of birds and one of the most important habitats  
150 of penguin, with a rich and extensive vegetation cover; Site Collins, located at the foot  
151 of the glacier Collins in an ice-free area about 10-15m from the sea.

152 Two more sites were chosen as intermediate: Sites Cerro Fosil and Cerro GPS, located  
153 on top of a small hill of about 25 m a.s.l., in the vicinity west of the Aerodrome T.R.  
154 Marsh, separated from it by no more than 100 m.

155

156 **Analytical techniques and methods**



157 *Magnetic techniques*

158 Magnetic measurements were carried out with two objectives: to quantify the magnetic  
159 minerals through measurements of magnetic susceptibility and remanent magnetization  
160 ( $M_r$ ), and to obtain magnetic parameters that might give information about the nature of  
161 magnetic minerals and their magnetic grain size.

162 In order to perform magnetic measurements, each sample of lichen and substrate was  
163 crushed into a powder and tightly packed into a gelcap. A lichen gelcap contains 150-  
164 200 mg of material, while a substrate gelcap contains between 300 and 400 mg of  
165 material. Then, the gelcaps were used for measurements with two different equipment.  
166 Firstly, the magnetic susceptibility, which is the ability of a sample to be magnetized in a  
167 weak field, was measured using an AGICO Kappabridge KLY-3 at the Institut de  
168 Physique du Globe de Paris (IPGP). The same samples were then used for the  
169 measurement of hysteresis parameters and First-Order Reversal Curve (FORC) diagrams  
170 with a Vibrating Sample Magnetometer (VSM) from Lakeshore, which is part of the  
171 Mineral Magnetism Analysis Platform situated of IPGP-IMPMC. More detail on the  
172 measurement of these properties can be found in the Supplementary Information.

173 *Scanning Electron Microscopy with X-ray Energy Dispersive Spectrometry*

174 This technique allows visualization of the external morphology of the sample. To obtain  
175 the image of the analysed surface, the sample is swept with an electron beam in a raster  
176 pattern. In the case of MEB-EDS, the backscattered electrons are used to generate the  
177 image. An X-ray detector is coupled to this system to perform an elemental qualitative  
178 and quantitative analysis of the sample.

179 The analysis was carried out in the microscopy laboratory of the ENES Morelia unit of  
180 the Universidad Nacional Autónoma de México (UNAM). Substrate and stem fragments  
181 from lichen samples were analysed using a JSM IT300 scanning electron microscope,  
182 JEOL brand with EDS X-Max detector with 20 mm<sup>2</sup> window. The substrate samples  
183 were metalized with a gold coating. The analysis was done under low vacuum  
184 conditions (50 Pa) for the lichen samples and high vacuum (10<sup>-4</sup> Pa) for the substrate  
185 samples. Elemental analysis using X-ray fluorescence (EDS) was done on some  
186 individual particles.

#### 187 *Instrumental Neutron Activation Analysis*

188 Instrumental Neutron Activation Analysis (INAA) is a multi-elemental and non-  
189 destructive technique that uses characteristic nuclear reactions. Neutron flux interacts  
190 with the sample, producing a nuclear reaction, mainly of the type (n,  $\gamma$ ). The  
191 characteristic gamma rays emitted by each radionuclide are captured by a high-  
192 resolution detector.

193 INAA technique have been used widely to study the elemental composition of samples  
194 from Antarctica (e.g. Mroz et al. 2018), whether they are sediments, lichens or mosses.  
195 In this work, we used the INAA technique to measure the concentrations of the elements  
196 Na, Sc, Cr, Fe, Co, As, La, Ce, Sm, Eu, Yb, Hf in the substrate of the sites LARC,  
197 Aerodromo and Ardley peninsula sampled at the same time than the lichens in the years  
198 1997 and 2006. Experimental details can be found in the SI.

199

200 *Elemental quantification by Inductive Coupled Plasma-Optical Emission Spectrometry*  
201 *(ICP-OES)*

202 We selected the elements to be quantified by Inductive Coupled Plasma-Optical Emission  
203 Spectrometry (ICP-OES) according to these 4 criteria: a) Potentially toxic chemical  
204 elements; b) Probability of finding them in the study area in concentrations quantifiable  
205 by the technique used, based on previous research; c) Bio accumulative chemical  
206 elements; d) Elements of known anthropic origin. Therefore, the quantified elements,  
207 both for lichens and for the substrate, were Ag, Al, As, B, Ba, Be, Cd, Co, Cr, Cu, Fe,  
208 Mn, Mo, Ni, Pb, Se, V and Zn. However, many of them were found under detection  
209 limits (UDL) of the technique. The experimental procedure is detailed in the SI section.

210

## 211 **Results**

### 212 *Magnetic measurements*

213 According to the Day-diagrams, hysteresis parameters for the lichen samples (1997 to  
214 2010) all plot in the pseudo-single-domain (PSD) region, between the single-domain  
215 (SD)- multidomain (MD) (MD percentage >60%) and the SD-superparamagnetic (SP)  
216 mixing lines (SP percentage >30%) (Fig.2a).

217 More than 3/4 of the  $M_r/M_s$  ratios are between 0.1 and 0.2, and 85% for the  $H_{cr}/H_c$  ratios  
218 are between 3 and 5. Most of the points that are not in this region are from Ardley site,  
219 and they are characterized by a smaller magnetic grain size, with points closer to the SD-  
220 SP mixing line, with a higher SP percentage. This tends to show that magnetic particles  
221 present on all the sites, except Ardley, have a similar origin. On the contrary, the  
222 hysteresis parameters from the substrate samples plot slightly closer to the SD-SP  
223 mixing line than the lichen samples, but they are still in the PSD region (Fig. 2b).

224 Samples from Ardley have a behaviour different from the rest of the samples, identically  
225 to what we observed in the lichens, and they plot closer to the SP region.

226 These results are further confirmed with the good correlation between magnetic  
227 susceptibility and  $M_r$  in the case of the lichen samples (Fig. 3a), where the determination  
228 coefficient  $R^2=0.889$  (95% significance level), but for the substrate samples, there is  
229 only little correlation between magnetic susceptibility and remanent magnetization  
230 ( $R^2=0.378$ ), showing that the magnetic sources in these samples are diverse (Fig. 3b).

231 The magnetic signal in the substrate is likely caused not only by the anthropogenic  
232 signal, but it also takes into account geological sources.

233 The variation of  $M_r$  with time shows an increase in  $M_r$  values in 1997 (when there is  
234 available data) and another one in 2006-2007(Fig. 4a). Susceptibility values show a  
235 large variation, from almost zero to  $2.5 \times 10^{-6}$  SI  $m^3/kg$ . The variation of magnetic  
236 susceptibility is similar, again with a peak in 1997 and 2006-2007 (Fig. 4b).

237 In order to study the spatial variation of magnetic parameters, we plotted the remanent  
238 magnetization values and the susceptibility values for lichen samples from 2010 on a  
239 cartographic representation of the sites (Fig. 5). That year corresponds to a sampling of  
240 all the sites and the results do not seem to be affected by the external causes that  
241 increased the magnetization and susceptibilities everywhere. The lowest values  
242 correspond to samples from the Collins ice sheet and Ardley peninsula, where there is  
243 little human activity. On the contrary, the sites with the highest magnetic susceptibility  
244 are the sites close to the airport and the site of Cerro Fósil in particular.

245 Unfortunately, the only sampling years for which we have substrate samples available  
246 for this study are 1997, 1998 and 2006, which are the years corresponding to the  
247 possible occurrences of external sources of magnetic input. It is therefore difficult to  
248 make any interpretation on the variations of magnetic parameters with time (Fig. SI-2).

249 *FORC diagrams- Comparison magnetic signal lichen/substrate*

250 Because they are calculated with a second derivative of magnetization, FORC diagrams  
251 are extremely sensitive to noise, and therefore can only be measured on the magnetically  
252 strongest samples. We measured FORC diagrams on the strongest lichen samples (most  
253 of them from the 1997 sampling) and on corresponding lichen and substrate samples  
254 whenever available.

255 Fig. 6 shows some of the FORC diagrams measured, over three different sampling years.  
256 Figs 6a., c., e. and g. are FORC diagrams measured on lichens, and Figs 6b., d., f. and h.  
257 show FORC diagrams measured on the substrate samples. The first three pairs were  
258 measured on corresponding lichen and substrate samples. Firstly, patterns characteristic  
259 of a SD-PSD magnetic grain size are visible on all FORC diagrams. The SD contribution  
260 is identified with the high coercivity tail, and the PSD contribution by the peak of FORC  
261 distribution close to the origin of the diagram and the fact that most contours do not  
262 close but instead they intersect the vertical axis. The spread along the vertical axis is  
263 quite limited, except for sample ALL97 (Lichen- LARC 1997), which shows that  
264 magnetic interactions are moderate.

265 *Elemental concentrations in the substrate determined by INAA and ICP-OES*

266 Figure 7 shows the elemental concentrations determined by INAA and ICP-OES of  
267 substrate from three of the sites sampled in the years 1997 and 2006.

268 These results are somewhat surprising since the continental terrestrial crust normally  
269 presents higher concentrations of Al than Fe. The higher concentrations of Na than Al  
270 agree with the marine aerosol contribution on King George Island.

271 Scandium, Yb and Eu are natural elements, which could explain the low variability of  
272 these elements on the different sites. In contrast, Cu, Zn, Co, Ni, Pb and As are  
273 anthropogenic elements, and therefore they show a great variability depending on the  
274 sites and years.

275 *Elemental concentrations determined by SEM-EDS*

276 Figure 8 shows examples of SEM micrographs and semi-quantitative EDS analysis  
277 corresponding to individual particles that contain heavy elements, and therefore could  
278 carry the magnetic signal as well as heavy metals, both in lichen and the substrate. SEM  
279 micrographs of lichens (Fig. 8a-b) show the presence of particles of a wide range of  
280 sizes (from sub-micrometres to several tens of micrometres), dispersed in the sample.

281 EDS analysis in lichens all show the presence of natural elements such as Si, Al, Mg.

282 Among the possible anthropogenic elements, Cr is present on two sites sampled in 1997  
283 (Collins and Base China). Small amounts of K, P and V are also measured in various  
284 sites and sampling years. Calcium is also present in small amounts but widely  
285 throughout the different sites.

286 Angular particles between 1-20  $\mu\text{m}$  can be seen on SEM micrographs of substrate  
287 samples (year 1997) (Fig.8c-d). EDS measurements also show the presence of the same

288 natural elements (Si, Al, Mg) that were identified in the lichen samples. Titanium is also  
289 present in the soil; the origin of this is probably natural. Other elements present in small  
290 amounts are Ca, V and K. These elements were also present in small amounts in the  
291 lichens.

292 *Elemental concentrations in lichens determined by ICP-OES*

293 Figure 9 shows the elements Fe, Al, Mn and Zn that are present with an important  
294 concentration (10-200 000 mg/kg), but different concentrations in the different sites. The  
295 concentrations of Zn in LARC site and Aerodromo site in 2006 are particularly high.  
296 Iron and Al concentrations are similar, and have the highest values in 2010, and the  
297 lowest in 2006, while Mn and Zn concentrations do not show such large variations.

298 The Fe concentrations measured here are above the USDA recommended maximum  
299 limits for *Usnea spp* (272 mg/kg) for all the years and sites except Collins and Ardley in  
300 1997 and Ardley 2006. Aluminium exceeds the recommended value (499 mg/kg) at  
301 LARC and Base China in 1997, Base China, Cerro Fosil and Ardley in 2006 and  
302 everywhere except Aerodromo MET in 2010.

303 Figure 10 shows the presence of 13 elements in concentrations between 0.01 to 5 mg/kg,  
304 in lichens of the different sites in 1997. The highest concentrations of Cu and Pb are  
305 measured at the sites closer to the airfield and bases (Cerro Fosil, Aerodromo, Base  
306 China), while Ardley and Collins sites have much lower values.

307 The same information for the years 2006 and 2010 is presented in the Supplementary  
308 Information (Figs. SI-3 and SI-4). Overall, the concentrations of Cr, Cd and Pb have a  
309 tendency to decrease over the years, but the concentration of Cu at the airfield shows a

310 strong increase in 2006. The lowest values are found at Ardley and Collins sites, the two  
311 sites most isolated from human activities.

312 Vanadium concentration is above the USDA recommended limit (1.5 mg / kg) at Base  
313 China, Aerodromo, Aerodromo MET and Cerro Fosil in 1997 and all sites except Ardley  
314 in 2006. Regarding Mo, the maximum recommended concentration is 0.40 mg/kg, and  
315 this value is exceeded at LARC, Aerodromo and Aerodromo MET in 1997, at LARC in  
316 2006 and everywhere except Aerodromo and Ardley in 2010. Beryllium concentration is  
317 0.06 mg/kg or less.

318

## 319 **Discussion**

### 320 *Magnetic measurements*

321 The average magnetic susceptibility in lichen samples for the year 2010 is  $7.5 \times 10^{-8}$   
322  $\text{m}^3/\text{kg}$ , and the average SIRM for that same year is around 1 to 10  $\text{mAm}^2/\text{kg}$ . These  
323 values are one to two orders of magnitude lower than the values measured on lichens  
324 from Argentinian cities by Chaparro et al. (2013), and about the same order of  
325 magnitude as values measured on lichens sampled close to a cement factory in Slovakia  
326 (Paoli et al. 2016). For the years 1997 and 2006-2007, when both susceptibilities and  
327 SIRM values are the highest, the SIRM values are comparable to the lowest values  
328 measured in these three studies, and susceptibilities are comparable to the highest  
329 values. However, it should be kept in mind that the lichen species are all different in all  
330 these studies so direct comparison with these previous results is difficult.



331 The cartography of magnetic parameters enables us to identify the sites that receive the  
332 most magnetic particles. Regardless of the sampling year, the highest values are  
333 measured for the sites that are close to the airstrip: either one of the three sites the closest  
334 to the airstrip, or the site Aerodromo MET, or either the site of Cerro Fosil or Cerro  
335 GPS. These two sites are slightly further from the airstrip but they are higher in altitude  
336 and therefore could be impacted by the emissions from planes during take-off or  
337 landing.

338 At the other side of the spectrum, the lichen samples from Collins ice-sheet and Ardley  
339 peninsula have a very low magnetic signal. They are almost not impacted by direct  
340 human activities because they are far from all the scientific bases. Furthermore,  
341 hysteresis parameters for samples from Ardley peninsula are different from other  
342 samples: on the Day-plot, they all plot closer to the SP mixing line and SD region. The  
343 Peninsula hosts a penguin colony, and this difference in hysteresis parameters could be  
344 linked to an incipient formation process of soil by organic matter accumulation and  
345 associated phosphatization (Machado et al. 2006). Actually, high levels of SP grains in  
346 soils are often due to magnetic enhancement by secondary magnetite/maghemite  
347 produced by pedogenic processes (Dearing et al. 1997).

348 The FORC diagrams measured in this study can be compared with those measured by  
349 Salo et al. (2012) on lichens from urban and industrial sites. Despite the low resolution  
350 of their measurements, it is easy to see that their FORC diagrams are much more  
351 characteristic of multidomain (MD) magnetic grains than ours are. The FORC diagrams  
352 measured by Sagnotti & Winkler (2012) on tree leaves from Rome are also MD-like and  
353 therefore quite different from the diagrams we measured.

354 The only published paleomagnetic study from Antarctic Peninsula also includes FORC  
355 diagrams (Gao et al. 2018). Measurements on basalt samples from three different  
356 formations show patterns that are MD-like, with no high-coercivity component and a  
357 large vertical spread. On the other hand, a FORC diagram measured on volcanoclastic  
358 rocks from Fildes Peninsula is SD-like. In any case, our FORC diagrams show patterns  
359 that are very different from those measured on rocks from the Antarctic Peninsula,  
360 which tends to indicate that the magnetic signal we have measured is not mostly caused  
361 by geological input.

362 Comparison of magnetic results between corresponding lichen and substrate samples  
363 show a few differences. The substrate samples from sites close to the airstrip still have  
364 higher values than the samples from the other sites, but unlike for lichen samples,  
365 substrate samples from LARC and Collins have high  $M_r$  and susceptibilities. FORC  
366 diagrams for lichen and substrate samples also bear some differences. Even though they  
367 bear similar SD-PSD patterns, there are two significant differences: the SD tail is more  
368 prominent and longer, and the spread along the vertical axis more limited for the  
369 substrate than for the lichen samples. This shows that the magnetic particles deposited  
370 on the substrate are different from those on the lichen. It is probable that the magnetic  
371 signal in the substrate samples contain a geological part as well as anthropogenic part.  
372 Another explanation could also be that coarser-grain particles from anthropogenic origin  
373 are more efficiently fixed on the lichens, than in the substrate. It also seems that magnetic  
374 particles are more dispersed in substrate than in lichens, perhaps because they become  
375 more strongly fixed when deposited on lichen than on substrate. These observations are  
376 further confirmed by the fact that there is no correlation between  $M_r$  or magnetic  
377 susceptibility on sister substrate/lichen samples (Fig. 11). This shows that the signal

378 from the lichen *U. aurantiaco-atra* is likely to be more representative of the  
379 anthropogenic contribution than the signal from the substrate.

380 *Comparison magnetic-elemental concentration measurements*

381 The concentrations of Fe determined by INAA in the substrate samples of 1997 from  
382 Aerodromo, LARC and Ardley sites are consistent with the saturation magnetization  
383 measurements: for both data sets, the highest values are observed for the Aerodromo  
384 site, then an intermediate value for LARC and a lower value for Ardley. Similarly, the  
385 increase of Fe concentration from 1997 to 2006 also corresponds to an increase of  
386 saturation magnetization. However, when looking at the concentrations of the other  
387 elements, only the Ni concentrations for the year 1997 show similar variations of  
388 concentrations among sites.

389 In the lichen samples, the concentrations of the various elements measured by ICP-OES  
390 can be compared with the magnetic proxies (saturation magnetization and magnetic  
391 susceptibility). The element that shows the highest correlation with magnetic parameters  
392 is Ni ( $R^2=0.832$  for  $M_r$  and  $0.6173$  for  $MS$ ).

393 Some elements show a weak correlation: Cr ( $R^2=0.334$  for  $M_r$  and  $0.349$  for  $MS$ ); Co  
394 ( $R^2=0.420$  for  $M_r$  and  $0.243$  for  $MS$ ); V ( $R^2=0.342$  for  $M_r$  and  $0.154$  for  $MS$ ) and Ag  
395 ( $R^2=0.088$  for  $M_r$  and  $0.254$  for  $MS$ ). The correlation for Cu is even weaker ( $R^2=0.069$   
396 for  $M_s$  and  $0.136$  for  $MS$ ). Nickel and Cr are elements for which an association with  
397 magnetic parameters has been described (e.g., Cao et al. 2015, Salo et al. 2016),  
398 however it is somewhat surprising that no stronger correlation is observed for the other  
399 elements such as Cu, Pb, Cd, Ba, Mo.

400 *Possible sources of the various elements*

401 Possible explanations for the presence of the various elements in sometimes fairly high  
402 concentration, both in the lichen and the substrate, are presented in Table II.

403

404 **Conclusion**

405 We carried out for the first time magnetic and elemental analysis on *Usnea aurantiaco-*  
406 *atra* (Jacq.) Bory lichen and its substrate from various sites on the Antarctic Peninsula  
407 with various levels of influence from human activities. The main results are the  
408 following:

- 409 - Several anthropogenic elements have been identified on sites close to human  
410 activities: Ni, Pb, Mo, Cd, and Zn are the most prevalent. Some of the elements  
411 were found in sites that should have very little human influence, which shows  
412 that elements coming from other continents can travel long distances and  
413 contribute to the pollution in Antarctica.
- 414 - Magnetic proxies from lichen show a spatial correlation with human influence  
415 (scientific bases or airstrips). The temporal variation shows that on average,  
416 magnetic parameters started with high values in 1997, then decreased with time  
417 until 2007 but increased again in 2006-2007. The most recent measurements  
418 indicated nonetheless a general decreasing trend.
- 419 - It seems that the magnetic signal from the lichen is likely to be more  
420 representative of the anthropogenic contribution than that from the substrate.
- 421 - We observed a correlation between magnetic parameters and Ni, and to a lesser  
422 extent with Cr, Co, V, Ag.

423 - All these results suggest that using magnetic and elemental techniques, it is  
424 possible to implement monitoring with lichen *Usnea aurantiaco-atra* (Jacq.)  
425 Bory as a bioindicator for anthropogenic pollution.

426

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429 support. They also thank the editor and two anonymous reviewers for their constructive  
430 comments.

431

#### 432 **Author contribution**

433 MP performed the fieldwork and supervised the entire work. RF, CE, NG and CC  
434 performed the different analysis of the samples. CC, MP, BA and NG interpreted the  
435 obtained results. CC and MP prepared the manuscript and created the figures with input  
436 from BA and NG.

437

#### 438 **Details of data deposit**

439 The article will be deposited with free access in the libraries of the Universidad de Chile,  
440 the Sorbonne Université and the Universidad Nacional Autónoma de México.

441

442

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581

## 582 **Figure captions**

583 Figure 1. Sample sites in Fildes peninsula, King George Island, and lichen *Usnea*  
584 *aurantiaco-ater*(Jacq.) Bory.

585 Figure 2. Day-plot showing the hysteresis parameters with the mixing lines of Dunlop  
586 (2002) for: a: lichen *Usnea aurantiaco-atra* (Jacq.) Bory samples; b: substrate samples.

587 Figure 3. Susceptibility vs remanent magnetization for: a: lichen *Usnea aurantiaco-atra*  
588 (Jacq.) Bory samples; b: substrate samples.

589 Figure 4. Variation of a: remanent magnetization; b: magnetic susceptibility of lichen  
590 *Usnea aurantiaco-atra* (Jacq.) Bory samples with time (1997-2010).

591 Figure 5. Cartography of: a: remanent magnetization; b: magnetic susceptibility for  
592 lichen *Usnea aurantiaco-atra* (Jacq.) Bory samples sampled in 2010.

593 Figure 6. FORC diagrams. Top row: *Usnea aurantiaco-ater*(Jacq.) Bory samples;  
594 bottom row: substrate samples. Horizontal and vertical scales are the same for all  
595 samples except AsAr-06.

596 Figure 7. Elemental concentrations determined by INAA (Fe, Na, Zn, Sc, Cr, Ce, Co,  
597 As, La, Hf, Sm, Yb and Eu, labeled with an asterisk) and ICP-OES (Al, Cu, Ba, Ni, Pb  
598 and Cd) of substrate from three of the sites sampled in the years 1997 and 2006.

599 Figure 8. Examples of representative micrographs and EDS semi-quantitative analysis  
600 for: a: lichen sampled at Collins site in 1997; b: lichen sampled at Base China site in  
601 1997; c: substrate sampled at Collins site in 1997; d: substrate sampled at Aerodromo  
602 MET site in 1997.

603 Figure 9. Concentrations of major elements determined by ICP-OES of *Usnea*  
604 *aurantiaco-ater*(Jacq.) Bory from the seven sites sampled in the years 1997, 2006 and  
605 2010.

606 Figure 10. Concentrations of 13 minor elements determined by ICP-OES of *Usnea*  
607 *aurantiaco-ater*(Jacq.) Bory from the seven sites sampled in the year 1997.

608 Figure. 11. a:  $M_r$  of substrate sample vs.  $M_r$  of the corresponding *Usnea aurantiaco-*  
609 *ater*(Jacq.) Bory sample (taken at the same location on the same year). b: Magnetic

610 susceptibility of substrate sample vs. magnetic susceptibility of the corresponding lichen

611 sample.

612

613 **Tables**

614 Table I. Coordinates of the sampling sites of lichens *Usnea aurantiaco-atra* (Jacq.) Bory  
615 and the corresponding substrate

<b>Sampling site</b>	<b>Coordinates</b>	
Aerodromo	-62°11'23.28"	-58°58'27.12"
Cerro Fossil	-62°12'18.72"	-58°58'23.16"
Cerro GPS	-62°11' 57.74"	-58°58'28.89"
Aerodromo MET	-62°11'20.76"	-58°58'43.68"
Base China	-62°12'51.84"	-58°57'47.52"
LARC	-62°12'07.56"	-58°57'39.96"
Ardley	-62°12'46.44"	-58°55'49.8"
Collins	-62°10'08.76"	-58°51'06.84"

616

617

618 Table II – Possible origins of the elements detected in lichen *Usnea aurantiaco-atra*  
 619 (*Jacq.*) *Bory* and the corresponding substrate samples

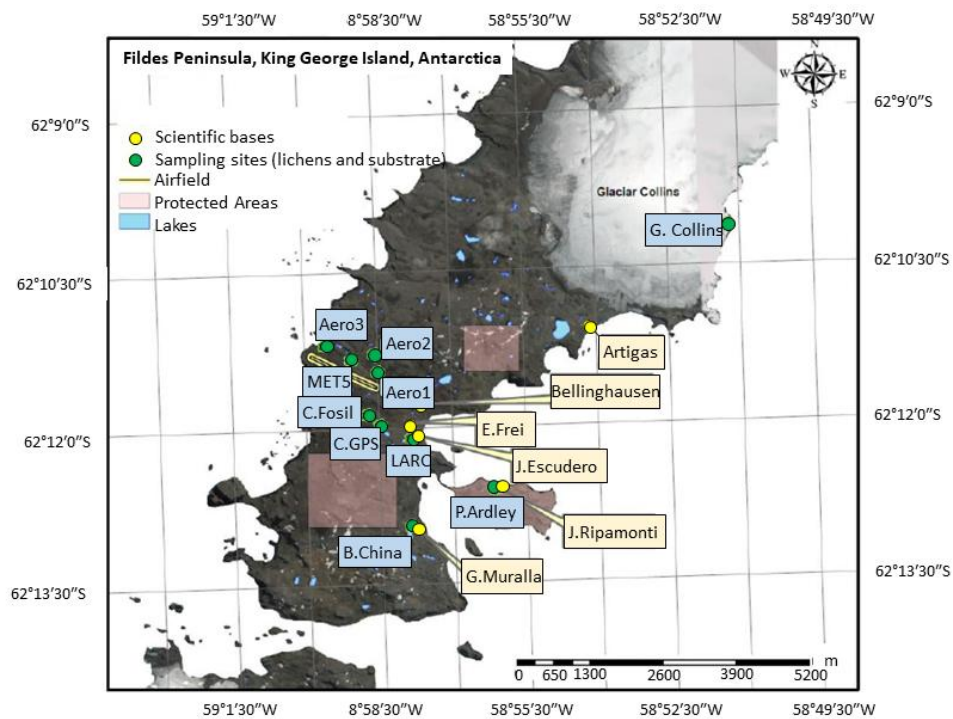
Element	Possible source
Fe, Al (2010)	Dust from road construction and extension of Chinese scientific base
Ni	Oil and diesel combustion(Pereira et al. 2010); use of lubricating oil (Song et al. 2010); additives in airplane fuel to increase motor efficiency (Marín-Flores et al. 2010); electric batteries
Pb	Airplane fuel burning (Pereira et al. 2010), abrasion of brake pads (Hong et al. 2020) and vehicle tires (Lough et al. 2005), oil lubricants (Song et al. 2010, Altıntaş et al. 2019); electric batteries
Cd	Elimination of wastes from incinerators (Boccaccini and Ondracek, 1995, Sekito et al. 2014); electric batteries
Mo	Elimination of wastes from incinerators (Boccaccini and Ondracek, 1995, Sekito et al. 2014); fossil fuel combustion (Pereira et al. 2010, Celo et al. 2015) and use of lubricating oil with molybdenite (Song et al. 2010) used in Antarctica in airplanes and machines such as snowploughs and construction machines; additives in airplane fuel (Marín-Flores et al. 2010)
Zn	Electric batteries, building construction work.
V	Use of diesel to produce electric energy (Hope 1997)
Be	Used with Cu or Ni alloys (Goddard et al. 2016)
La, Hf, Sm (Ardley)	Increasing use of these elements in catalytic systems of the new motor vehicles used in all other continents and arriving to Antarctica.
P (Ardley)	Presence of the penguin colony: the substrate is formed by organic matter accumulation and associated phosphatization (Machado et al. 2006).
Cr (Collins, 2006)	Extra-Antarctic source*

620 \*The earth crust and rocks contain about 100 mg/kg of Cr. Soils contain normally on the  
 621 average about 40 ppm of Cr, sometimes up to some hundreds mg/kg. (Merian 1984). In this  
 622 work we find around 20 mg/kg in substrate. Some plants can take up higher Cr amounts, for  
 623 instance lichens, mosses, ferns, grasses and others. We find lower concentrations of Cr in the  
 624 lichen. On the other hand, practically all chromium compounds used by man originate from  
 625 chromite, of which largest resources are placed in South Africa, Zimbabwe and Russia.

626



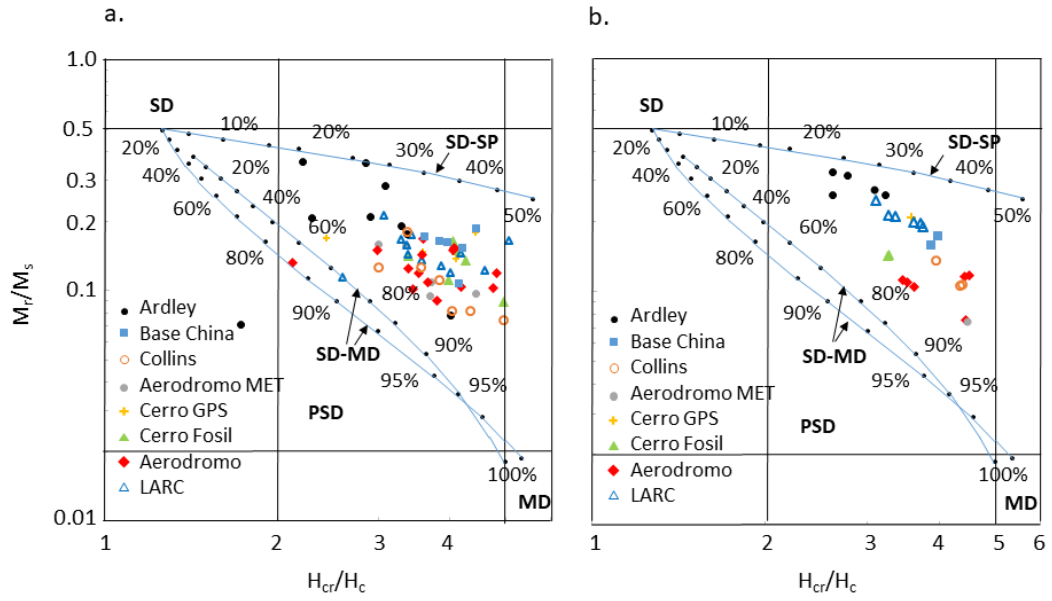
627 **Figures**



628

629 **Figure 1**

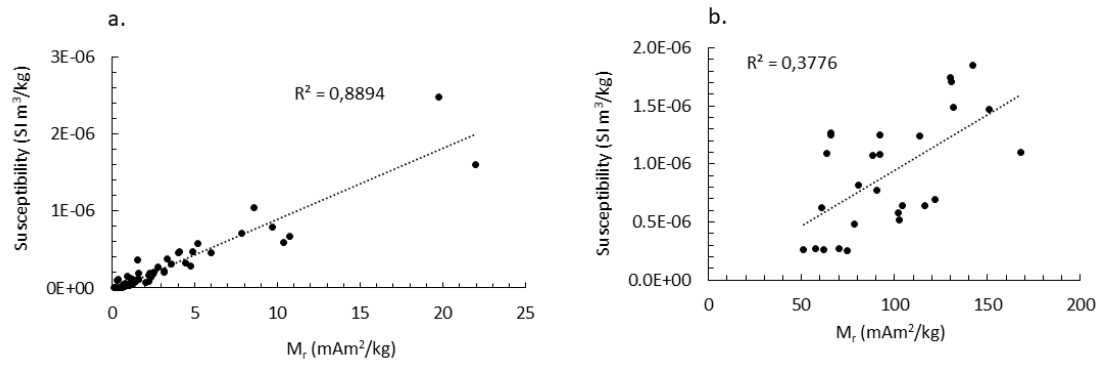
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632 Figure 2

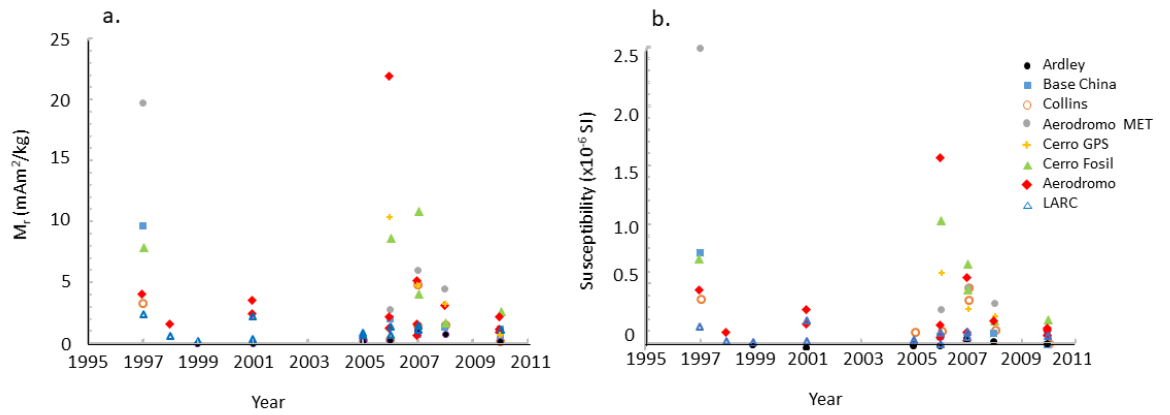
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635 Figure 3

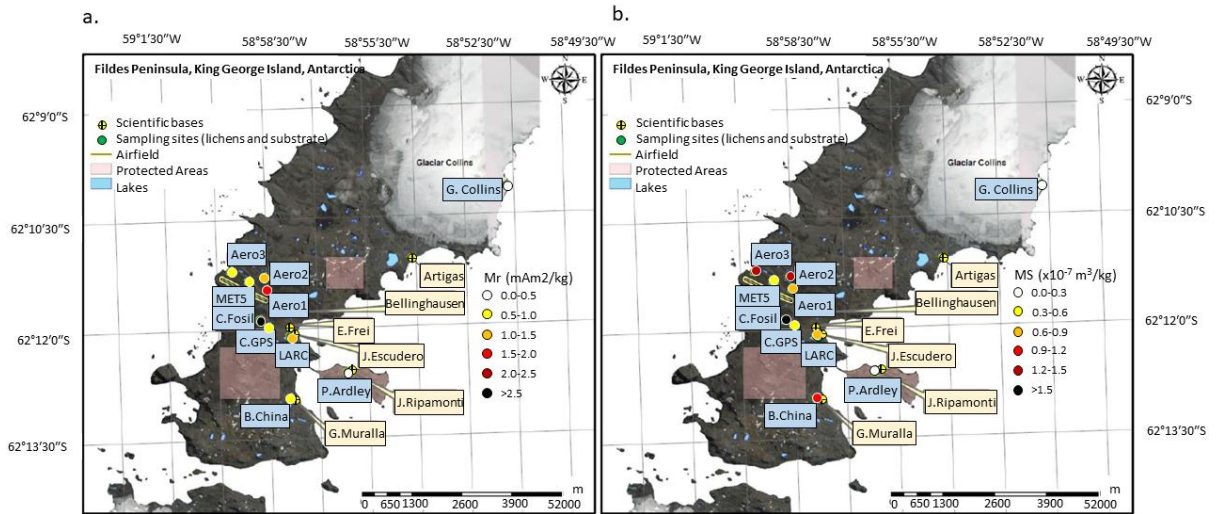
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638 Figure 4

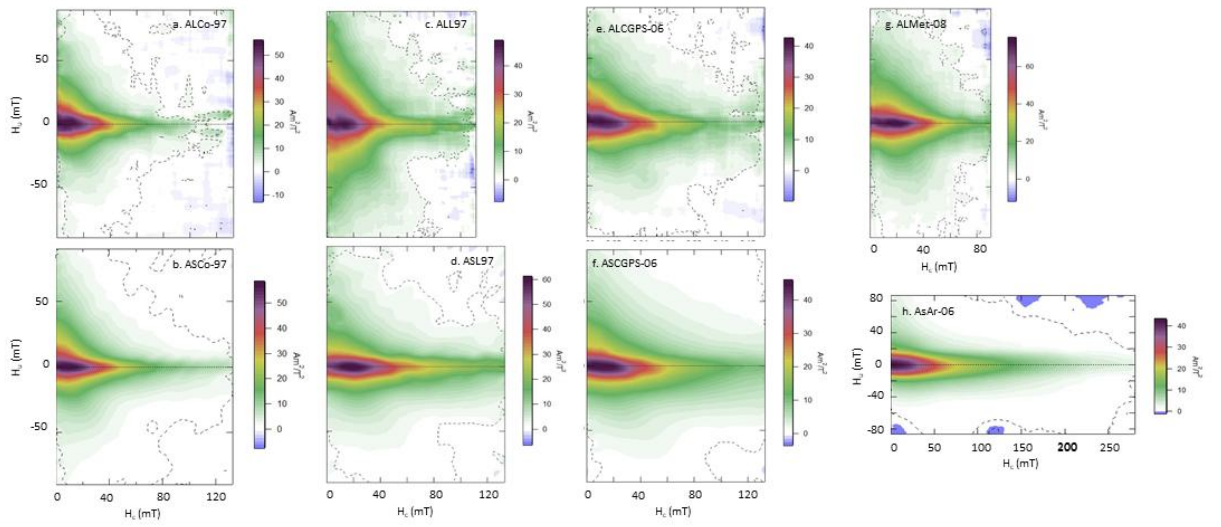
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641 Figure 5

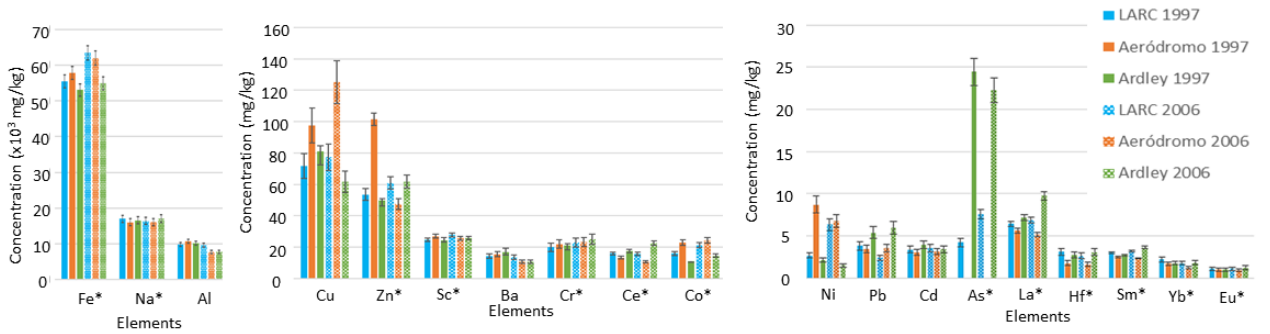
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644 Figure 6

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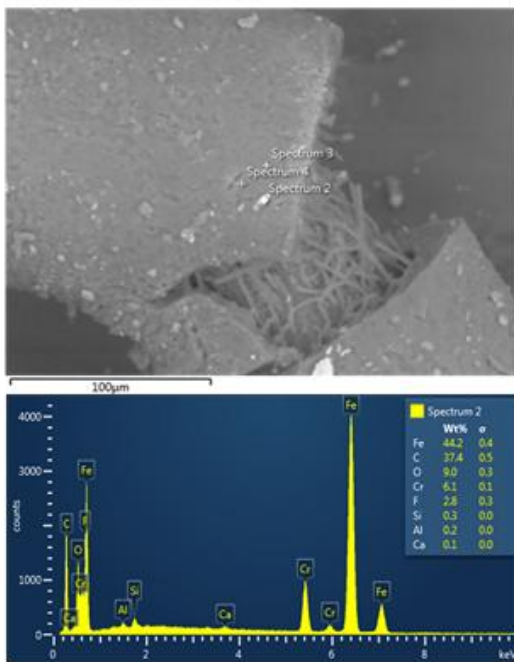


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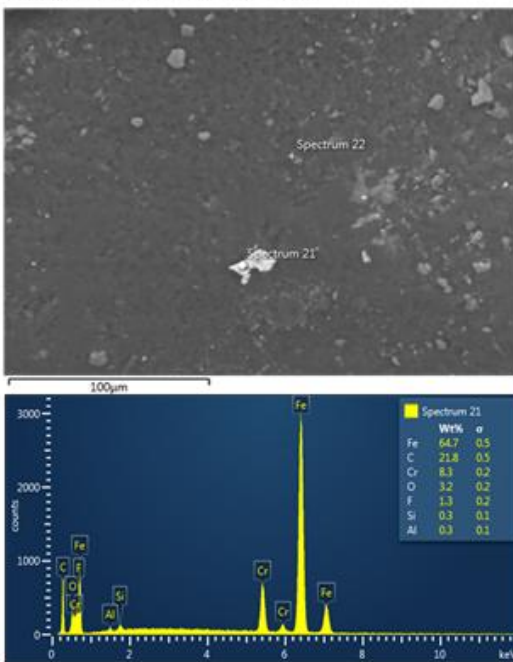
647 Figure 7

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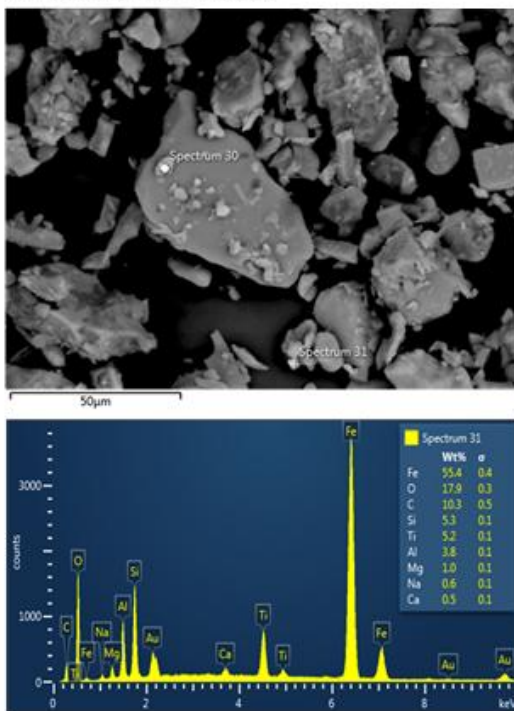
a. Lichen - Collins 1997



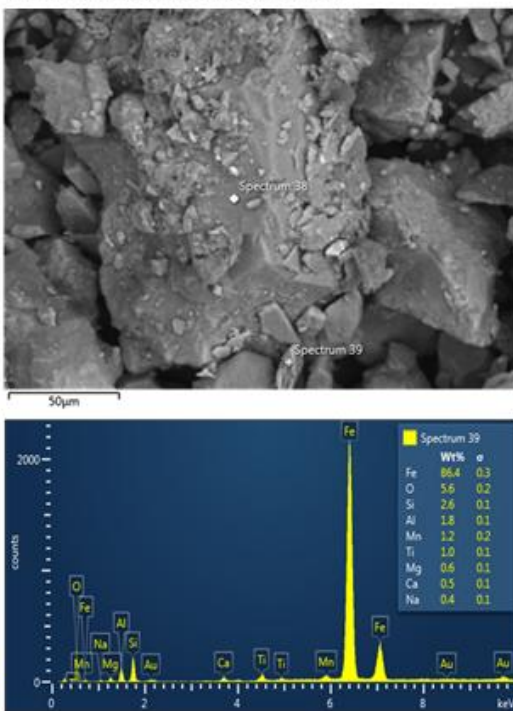
b. Lichen - Base China 1997



c. Substrate Collins 1997

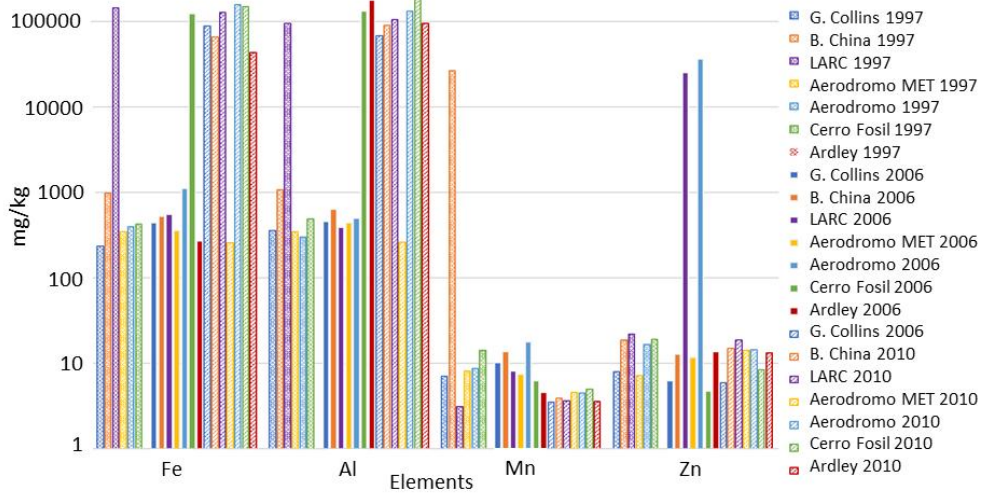


c. Substrate MET Airfield 1997



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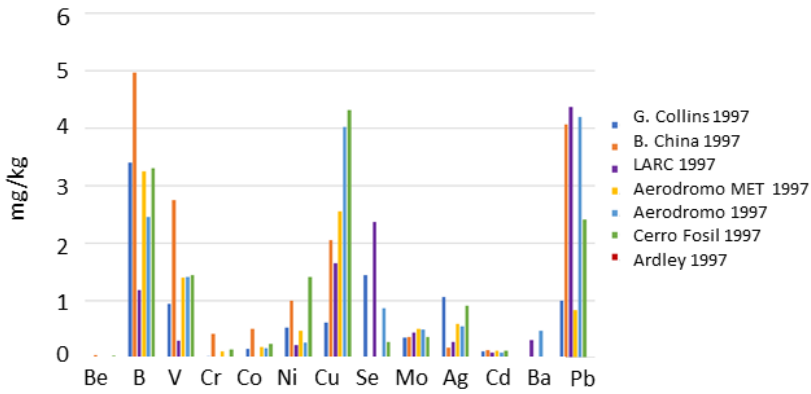
650 Figure 8



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652 Figure 9

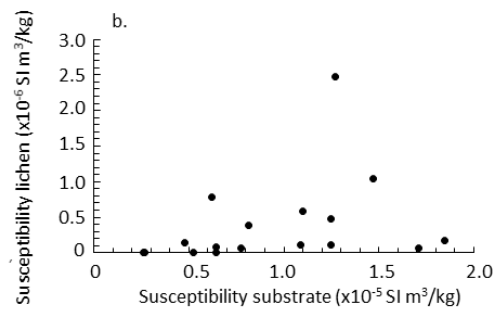
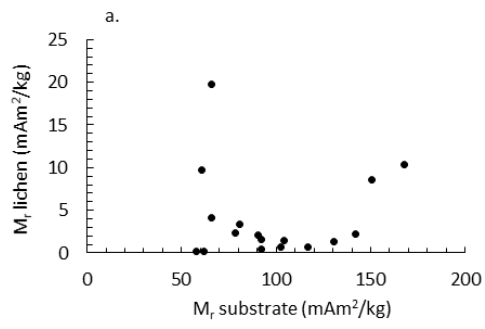
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655 Figure 10

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658 Figure 11