Fractional solubility of iron in mineral dust aerosols over coastal Namibia: a link with marine biogenic emissions?

Karine Desboeufs[1], Paola Formenti[1], Raquel Torres-Sánchez[2,3], Kerstin Schepanski[4,$], Jean-Pierre Chaboureau[5], Hendrik Andersen[6,7], Jan Cermak[6,7], Stefanie Feuerstein[4], Benoit Laurent[1], Danitza Klopper[8,*], Andreas Namwoonde[9], Mathieu Cazaunau[2], Servanne Chevaillier[2], Anaïs Feron[1,%], Cécile Mirande-Bret[1], Sylvain Triquet[1], and Stuart J. Piketh[8]

[1] Université Paris Cité and Université Paris Est Créteil, CNRS, LISA, F-75013 Paris, France

[2] Université Paris Est Créteil and Université Paris Cité, CNRS, LISA, F-94010 Créteil, France

[3] CIQSO, Robert H. Grubbs Building, University of Huelva, Campus El Carmen, E21071 Huelva, Spain

[4] TROPOS, Leipzig, Germany

[5] Laboratoire d'Aérologie (LAERO), Université de Toulouse, CNRS, UT3, IRD Toulouse, France

[6] Institute of Meteorology and Climate Research, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany

[7] Institute of Photogrammetry and Remote Sensing, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany

[8] North-West University, School for Geo- and Spatial Sciences, Potchefstroom, South Africa

[9] SANUMARC, University of Namibia, Henties Bay, Namibia

* Now at University of Limpopo, Department of Geography and Environmental Studies, Sovenga, South Africa

$ Now Institute of Meteorology, Freie Universität Berlin, Berlin, Germany

% Now at Université Paris-Saclay, INRAE, AgroParisTech, UMR ECOSYS, Palaiseau, France

Corresponding author: paola.formenti@lisa.ipsl.fr

Abstract

~~Mineral dust is the largest contributor to elemental iron in the atmosphere, and, by deposition, to the oceans, where elemental iron is the main growth limiting nutrient. Southern Africa is an important source at the regional scale and for the Southern Ocean, however limited knowledge is currently available about the fractional solubility of iron from those sources, as well as on the atmospheric processes conditioning its dissolution during deposition.~~

This paper presents the first investigation of the solubility of iron in mineral dust aerosols ~~from 176 filter samples~~ collected at the Henties Bay Aerosol Observatory (HBAO), in Namibia, from April to December 2017. During the study period, 10 intense dust events occurred. Elemental iron reached peak concentrations as high as 1.5 µg m^{-3}, significantly higher than background levels. These events are attributed to wind erosion of natural soils from the surrounding gravel plains of the Namib desert. The composition of the sampled dust is found to be overall similar to that of aerosols from northern Africa, but characterised by persistent and high concentrations of fluorine, which are attributed to local fugitive dust ~~from mining activities and soil labouring for construction~~.

The fractional solubility of Fe (%SFe) for both the identified dust episodes and background conditions ranged between 1.3 to 20 % and averaged at 7.9% (± 4.1%) and 6.8 (± 3.3%), respectively. Even in background conditions, the %SFe ~~iron fractional solubility~~ was correlated to that of ~~aluminium~~ Al and ~~Si~~silicon. The solubility was lower between June and August, and increased from September onwards, during the austral spring ~~months~~. The relation with measured concentrations of particulate MSA (methane sulfonic acid), solar irradiance and wind speed suggests a possible two-way interaction whereby marine biogenic emissions from the coastal Benguela upwelling to the atmosphere would increase the solubility of iron-bearing dust, according to the photo-reduction processes ~~proposed by Johansen and Key (2006). The subsequent deposition of soluble iron could act to further enhance marine biogenic emissions.~~ This first investigation points to the west coast of southern Africa as a complex ~~and dynamic~~ environment with multiple processes and active exchanges between the atmosphere and the Atlantic Ocean, requiring further research.

Keywords: aerosols, mineral dust, water-soluble Fe, atmospheric processing, marine biogenic emissions

1.   Introduction

Through the processes of atmospheric transport and deposition, mineral dust is known to provide nutrients and metals to the terrestrial and marine ecosystems (Hooper et al., 2019; Ventura et al., 2021). Amongst those, mineral dust provides iron (Jickells et al., 2005), which plays a major role for the primary productivity of the nutrient-limited oceans, modulating the marine carbon cycle (Hooper et al., 2019) as well as that of key continental ecosystems such as the Amazon rainforest (Reichholf, 1986).

To date, much attention has been paid to the soluble Fe in mineral dust emitted from arid and semi-arid areas in the northern Hemisphere, in particular the Saharan and Chinese deserts (e.g. Baker et al., 2006; Paris et al., 2010; Takahashi et al., 2011; Rodriguez et al., 2021), where emissions are the most intense (Tegen and Schepanski, 2009). Nonetheless, the southern Hemisphere accounts for approximately 10% of the global atmospheric dust loading (Kok et al., 2017). Large sources are found in southern Africa, mostly in Namibia (Kalahari and Namib deserts, Etosha Pan, numerous ephemeral riverbeds along the Namibian coastline) and Botswana (Makgadikgadi Pan) (Prospero et al., 2002; Bryant et al., 2007; Mahowald et al., 2003; Ginoux et al., 2012; Vickery and Eckardt, 2013; Von Holdt et al., 2017).

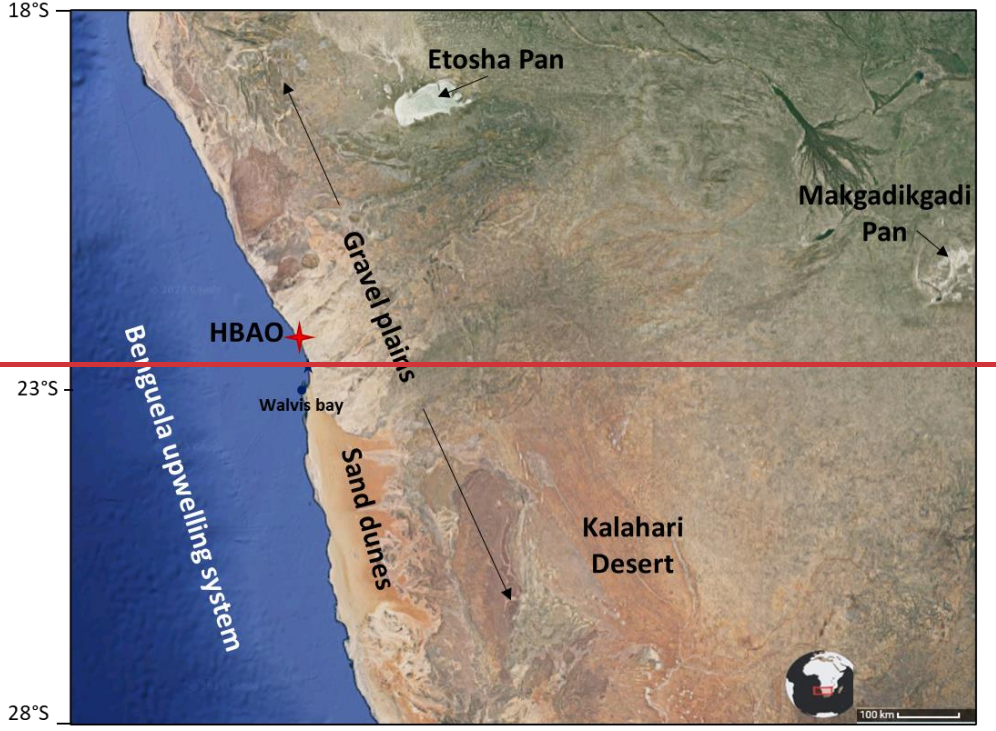
Previous research has shown that the long-range transport of dust emitted from southern African sources can reach the south-eastern Atlantic and the Indian Oceans (Swap et al., 1996; Jickells et al., 2005; Bhattachan et al., 2012; 2015; Ito and Kok, 2017). In particular, Gili et al. (2022) demonstrated recently that mineral dust from Namibia can also be transported across the Southern Oceans to eastern Antarctica. Furthermore, the research by Dansie et al. (2022) has suggested that mineral dust from Namibia could dominate the atmospheric deposition to the coastal Benguela Upwelling System (BUS), where biomass burning aerosols, a significant source of soluble Fe to the Southern and Indian Oceans (Hamilton et al., 2021; Ito et al., 2021; Liu, et al., 2022), are limited by atmospheric stratification (Formenti et al., 2019; Redemann et al., 2021). The inputs of Namibian (and Angola) dust in the upwelled waters could also modulate the migration of skipjack tuna between the Gulf of Guinea and the equatorial Atlantic, by contributing to support phytoplankton growth and hence upper trophic levels in this area (Rodriguez et al., 2023).

There is, however, very little data available on the concentrations and composition of soluble Fe in dust aerosols from southern Africa, both near the sources and over the oceans. Previous research in Namibia focussed on soils and sediments (Dansie et al., 2017a; 2017b; Kangueehi, 2021). The Atlantic Meridional Transect (AMT) cruise programme conducted recurrent observations between October and March in the South Atlantic Ocean (Baker et al., 2013), while Heimburger et al. (2013) and Gao et al. (2013) report on sparse measurements of deposited aerosols and in rainwaters over the Southern Indian Ocean.

Within this context, this paper investigates the fractional solubility of Fe (%SFe) in samples of atmospheric aerosol particles smaller than 10 µm in diameter collected in 2017 at the Henties Bay Aerosols Observatory (HBAO; 22.09°S, 14.26°E) on the Namibian coast. In section 2 we outline the experimental and analytical methodology for elemental and water-soluble analysis of ions and metals, including iron. We also provide the definition of fractional solubility and the method for estimating the total dust mass. We introduce the supporting tools used to evaluate the source regions of the collected mineral dust, their pathways during transport, and the presence of fog, a recurrent feature on coastal Namibia favouring multi-phase ageing processes. Section 3 provides the results of the analysis. We present the iron soluble concentrations and solubility, and explore their links to the load, emission area and transport of mineral dust, as well as atmospheric processing. Section 4 discusses the observations, suggesting that the %SFe fractional solubility of iron in the Namibian dust is higher when the MSA (methane sulfonic acid), a tracer of marine biogenic emissions, is also detected in these highest concentrations. This points to the photo-oxidation of DMS (Dimethyl Sulfide) as a process for increasing the Fe solubility, and suggests a possible positive feedback loop of the iron fertilisation by dust to the ocean. Section 5 summarizes the findings and suggests directions for future research.

2. Methodology

2.1. Study area

The Henties Bay Aerosol Observatory (HBAO, (22.09°S, 14.26°E; http://www.hbao.cnrs.fr/, last access: 10 October 2022) is located at the Sam Nujoma Marine and Coastal Resources Research Centre (SANUMARC) of the University of Namibia in Henties Bay, Namibia (Fig. 1).

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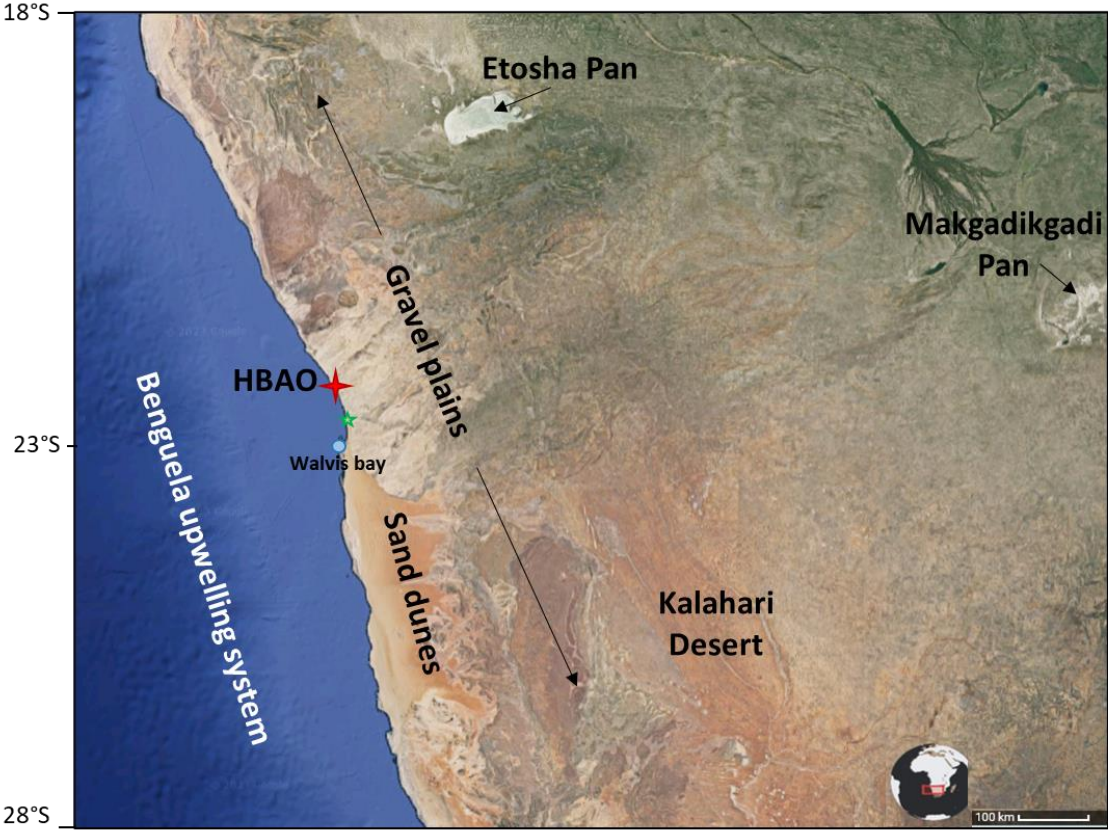
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116 **Figure 1: Location of Henties Bay Aerosol Observatory (HBAO, red star) and main dust source regions**
117 **(© Google Maps). The position of Walvis Bay (blue dot), the major harbour in the area, and the Wlotzkas-**
118 **baken meteorological station (~~blue~~ green star) are also indicated.**

119

120 Three kilometers to the south of the University campus hosting HBAO is the small town of Henties
121 Bay, with no industrial activity and very little traffic, and approximately 170 km north from Walvis Bay,
122 the major harbour in Namibia. Directly east of HBAO are the Namibian gravel plains, which are one
123 of the dominant features of the Namib desert together with the sand dunes. Approximately 100 m to
124 the north is the Omaruru riverbed, one of the coastal sources of mineral dust identified by Vickery and
125 Eckardt (2013).

126 Our previous results show that, at the surface level, the atmosphere at HBAO is a receptor of different
127 air masses dominated by marine aerosols, but also the seasonal occurrence of light-absorbing aero-
128 sols from biomass burning or pollution in northern wind regimes, and mineral dust detected episodi-
129 cally from various wind directions (Formenti et al., 2018; Klopper et al., 2020, hereafter KL20).

130 **2.2. Sample collection and analysis**

131 Aerosol particles smaller than 10 µm in aerodynamic diameter (PM_{10}) were collected by an automated
132 sampler (model Partisol Plus 2025i, Thermo Fisher Scientific, Waltham, MA USA) on 47 mm What-
133 man Nuclepore polycarbonate filters (1-µm pore size). The air was drawn through a certified sampling
134 inlet (Rupprecht and Patashnick, Albany, New York, USA) located at approximately 30 m above
135 ground and operated at a flow rate of 1 m^3 h^{-1}. Samples were collected for 9 hours during the daytime
136 (from 9:00 to 18:00 UTC time) and night-time (21:00 to 06:00 UTC time) for 12 non-consecutive weeks
137 from April to December 2017 (7-14 April, 26 April-3 May, 19-26 May, 07-14 July, 2-9 August, 15-22
138 August, 18-25 September, 02-09 October, 31 October-7 November, 13-20 November, 28 November-
139 04 December, 12-19 December). In total, 176 samples (+ ~~-~~13 blanks, one per week of sampling) were
140 collected.

141 The elemental analysis of 24 elements from Na to Pb, ~~and~~ including some major tracers of mineral
142 dust (Fe, Al and Si), was performed at the LISA laboratory by Wavelength-dispersive X-ray fluores-
143 cence (WD-XRF) using a PW-2404 spectrometer (Panalytical, Almelo, Netherlands), as detailed by
144 KL20. The total mass concentration per element x will be referred to as TX.

145 The measured elemental concentrations are used to calculate the estimated dust mass (*EDM*) ac-
146 cording to Lide (1992) as

147 EDM = 1.12 x {1.658 x [nss-Mg] + 1.889 x[Al] + 2.139 x [Si] + 1.399 x [nss-Ca] + 1.668 x [Ti] + 1.582
148 $\qquad\qquad\qquad$ x [Mn] + (0.5 x 1.286 + 0.5 x 1.429 + 0.47 x 1.204) x [Fe]} $\qquad$ (1)

149

150 where, as explained by KL20, nss-Mg and nss-Ca represent the non-sea salt fractions of Mg and Ca,
151 respectively.

152 The analysis of the water-soluble fraction was also performed at LISA. Individual filters were placed
153 in 20 mL of ultrapure water (MilliQ® *18.2* MΩ.cm) for 30 minutes. The solution was filtered (Nuclepore

154 polycarbonate filters with 0.2μm pore size) then divided into two sub-samples. One half was analysed

155 by Ion Chromatography (IC) using a Metrohm IC 850 device equipped with a column MetrosepA supp

156 7 (250/4.0 mm) for anions and with a Metrosep C4 (250/4.0 mm) for cations. The IC analysis provided

157 the concentrations of the following water-soluble ions: F^-, Cl^-, NO_3^-, SO_4^{2-}, formate, acetate, oxalate,

158 MSA^- (methanesulfonic acid), Na^+, NH_4^+, K^+, Ca^{2+} and Mg^{2+}. A calibration with certified standard multi-

159 ions solutions of concentrations ranging from 5 to 5000 ppb was performed and the uncertainty of the

160 analysis was estimated to be 5% (KL20).

161 The second half of the solution was acidified to 1% with ultrapure nitric acid (HNO_3) and analysed by

162 Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) using a Spectro ARCOS

163 Ametek® ICP-AES and by High-resolution Inductively Coupled Plasma-Mass Spectrometry (HR-ICP-

164 MS) using a Neptune Plus™ instrument by Thermo Scientific™. The calibration curve was performed

165 using standard multi-element solutions ranging from 2 to 1000 ppb for ICP-AES and 1 to 1000 ppt for

166 HR-ICP-MS (Desboeufs et al., 2022). These analyses provided the dissolved mass concentrations

167 (DX) of 25 water-soluble metals and metalloids, including Fe, Al, and Si. All sample concentrations

168 were corrected using the filter blanks for each sampling period.

169 Based on those analyses, the fractional solubility (%SX) representing the percentage solubility value

170 was calculated as

171

172 $$\%SX = 100 \times DX/TX \tag{2}$$

173

174 with DX and TX, the dissolved and total elemental concentration respectively.

175 Here, a leaching protocol using ultrapure water (UPW) was used to simulate wet deposition of parti-

176 cles, since the wet deposition dominates the total iron supply in the Southern Atlantic Ocean (Chance

177 et al., 2015). Moreover, the UPW leach enables the chemical reaction between iron with organic or

178 inorganic ligands, naturally dissolved from the particulate aerosols into rain droplets. However, it is

179 known that the extraction protocol modulates dissolution process and hence the values of %SF~~iron~~

180 ~~fractional solubility~~, in particular the estimates using UPW are higher in comparison to these one using

181 seawater, but lower than the acidic, buffered or reduction agent leach (Perron et al., 2020).

182 **2.3. Ancillary data**

183 Maps of the emission fluxes of mineral dust were calculated using the dust emission model described

184 by Feuerstein and Schepanski (2019), driven with hourly 10m wind fields at a 0.1° x 0.1° grid from

185 the European Centre for Medium-range Weather Forecasts (ECMWF). The dust emission parame-

186 terisation follows Marticorena and Bergametti (1995). Additional information on the soil type was taken

from the ISRC soil data set (FAO/IIASA/ISRIC/ISSCAS/JRC, 2012) and information on the aerodynamic roughness length was obtained from POLDER/ADEOS surface products following the works of Marticorena et al. (2004) and Laurent et al. (2005). The MODIS monthly vegetation product (MYD13A3 v6) was used to describe the vegetation cover, while the vegetation type was defined using the BIOME4 database (Kaplan et al., 2003). We additionally differentiated between different dust source types (alluvial fines, dunes and sand sheets) which allowed us to reflect the source diversity over Namibia and thus the spatial diversity in the soil's susceptibility to wind erosion. This layer was compiled following Feuerstein and Schepanski (2019) using MODIS surface reflectance (MOD09A1 v6). A MODIS retrieved map on surface water cover was used to eliminate flooded areas as active dust sources.

Back-trajectories of the air masses during the dust events were calculated from Meso-NH model (version 5.3). The model set-up is similar to the one used for the AErosols, RadiatiOn and CLOuds in southern Africa (AEROCLO-sA) field campaign (Formenti et al. 2019) and related case studies (Flamant et al. 2022; Chaboureau et al. 2022). In short, the model was run on a 5 km grid covering the southern tip of Africa and 67 stretched levels spaced by 60 m close to the surface and 600 m at high altitude. Meso-NH was run for 24 h for each dust event using initial and boundary conditions provided by the ECMWF operational analysis. Emission, transport and deposition of dust is described by the scheme of Grini et al. (2006). Back trajectories were computed online using three passive tracers initialized with the 3D-field of their initial conditions. Further details on the dust prognostic scheme, the backward trajectories and the physical parameterizations are given in Chaboureau et al. (2022).

The presence of fog and low clouds (FLC) along the Namibian coastline during dust events was analysed using an existing satellite-based fog and low-cloud data set (Andersen et al., 2019). The FLC detection algorithm used to create this data set was developed and validated specifically for this region. The algorithm is based on infrared observations from the Spinning Enhanced Visible and Infrared Imager (SEVIRI) aboard the geostationary Meteosat Second Generation (MSG) satellites, making use of both spectral and textural information. The FLC product is available at the native spatial and temporal resolutions of the SEVIRI sensor (3 km nadir, every 15 minutes), as described in Andersen and Cermak (2018). The FLC product does not specifically distinguish between fog and low clouds but captures the coastal boundary-layer cloud regime typical for the region and at HBAO that could interact with mineral dust. It has been shown to be consistent with synoptic-scale atmospheric dynamics (Andersen et al. 2020). The FLC data are used to calculate maps of average fog and low cloud coverage for the time periods of all dust events given in Table 1.

Observations of the local meteorology, including measurements of air temperature, relative humidity and fog, at the nearby Wlotzkasbaken meteorological station (22.31°S, 14.45°E, 73 m asl, see Fig.

221   1) part of the Southern African Science Service Centre for Climate Change and Adaptive Land Man-
222   agement (SASSCAL) ObservationNet (https://www.sasscal.org/; last accessed 14/04/2023), are
223   used.

224

3. Results

3.1.   Description of the dust episodes

The dataset discussed in this paper is based on 176 aerosol samples collected at HBAO, 42 of which were associated with 10 dust episodes. As detailed by KL20, events of mineral dust were identified as peaks in the time series of the mass concentrations of Al and non-sea-salt Ca^{2+} (nss-Ca^{2+}). The dust episodes investigated in this study are a subset of those presented by KL20, we therefore use their naming convention to facilitate the connections between the two papers (Table 1). In the following, we refer to samples collected during the dust episodes as "dust". Samples collected outside the dust events will be indicated as "background".

Table 1. Dates of dust events identified at HBAO from May to December 2017, following KL20. The number of samples collected during each episode is indicated in the column called "N". The average air temperature, relative humidity, wind speed and direction recorded at the nearby meteorological station in Wlotzkasbaken are reported. The maxima wind speed and corresponding direction are indicated in brackets in the corresponding columns. The average EDM is reported with in brackets the maximum of EDM during the event.

Episode identifier	Start and end date (UTC)	N	Air temperature (°C)	RH (%)	Wind speed (m s^{-1})	Wind direction (degN)	EDM (µg m^{-3})
Dust 04	19/05 09h – 20/05 18h	3	17.7	73.7	2.7 (6.2)	186 (185)	13 (14)
Dust 05	24/05 21h – 26/05 09h	3	18.1	63.3	2.3 (6.3)	183 (188)	21 (42)
Dust 06	11/07 09h – 13/07 09h	4	13.2	82.9	1.2 (5.4)	235 (193)	27 (45)
Dust 07	04/08 21h – 06/08 09h	4	12.5	87.0	1.2 (5.4)	233 (201)	10 (16)
Dust 08	17/08 21h – 19/08 09 h	4	11.9	80.6	1.3 (4.6)	324(129)	18 (21)
Dust 09	23/09 21h – 24/09 18h	2	15.6	84.3	3.1 (6.2)	309 (330)	11 (17)
Dust 10	05/10 21h – 08/10 09h	8	14.0	74.6	2.1 (5.9)	249 (228)	14 (23)
Dust 11	15/11 09h – 18/11 09h	6	16.7	66.1	3.2 (11.7)	231 (232)	31 (56)
Dust 12	30/11 09h – 01/12 18h	3	16.7	78.1	1.9 (5.7)	244 (195)	2 (3)
Dust 13	15/12 09h – 19/12 09h	7	16.9	76.9	2.9 (6.5)	252 (238)	10 (19)

240

The dust episodes were long-lasting (generally a few days). The dynamic of the emissive areas, air mass transport and fog coverage during the episodes (Fig. 2 and S1) is driven by the synoptic circulation, which, in southern Africa, is primarily affected by the high-pressure belt under the descending limb of the Hadley cell (Tyson and Preston-Whyte, 2014). The maps of dust emission fluxes and the air mass back-trajectories reflect this seasonality. During the first part of the year (episodes Dust 04

246   to 05), dust emissions originated from the gravel plains and the Etosha pan north of HBAO. During

247   this time of the year the transport to HBAO below 300 m asl was north- to south-easterly originating

248   inland from the coast.

249

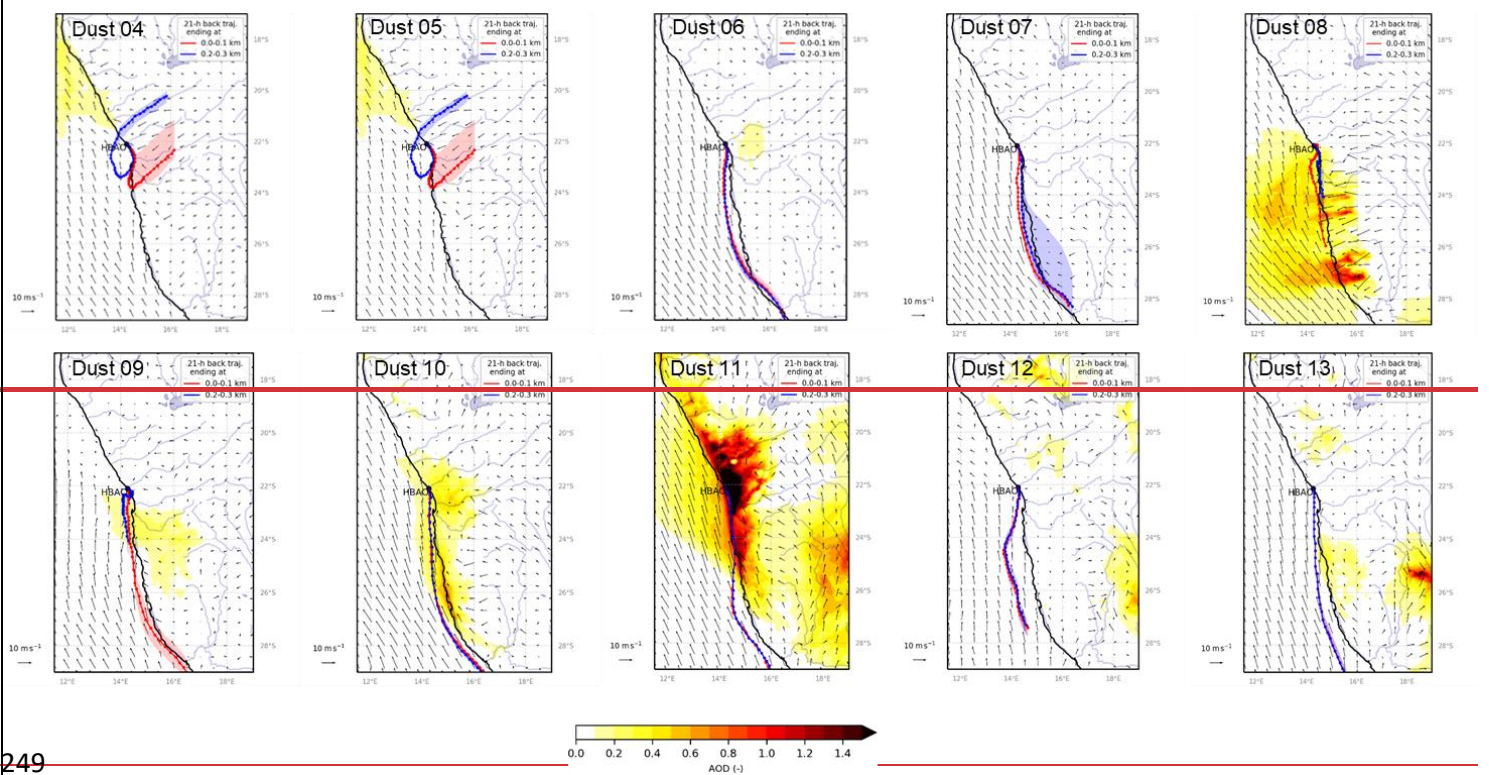
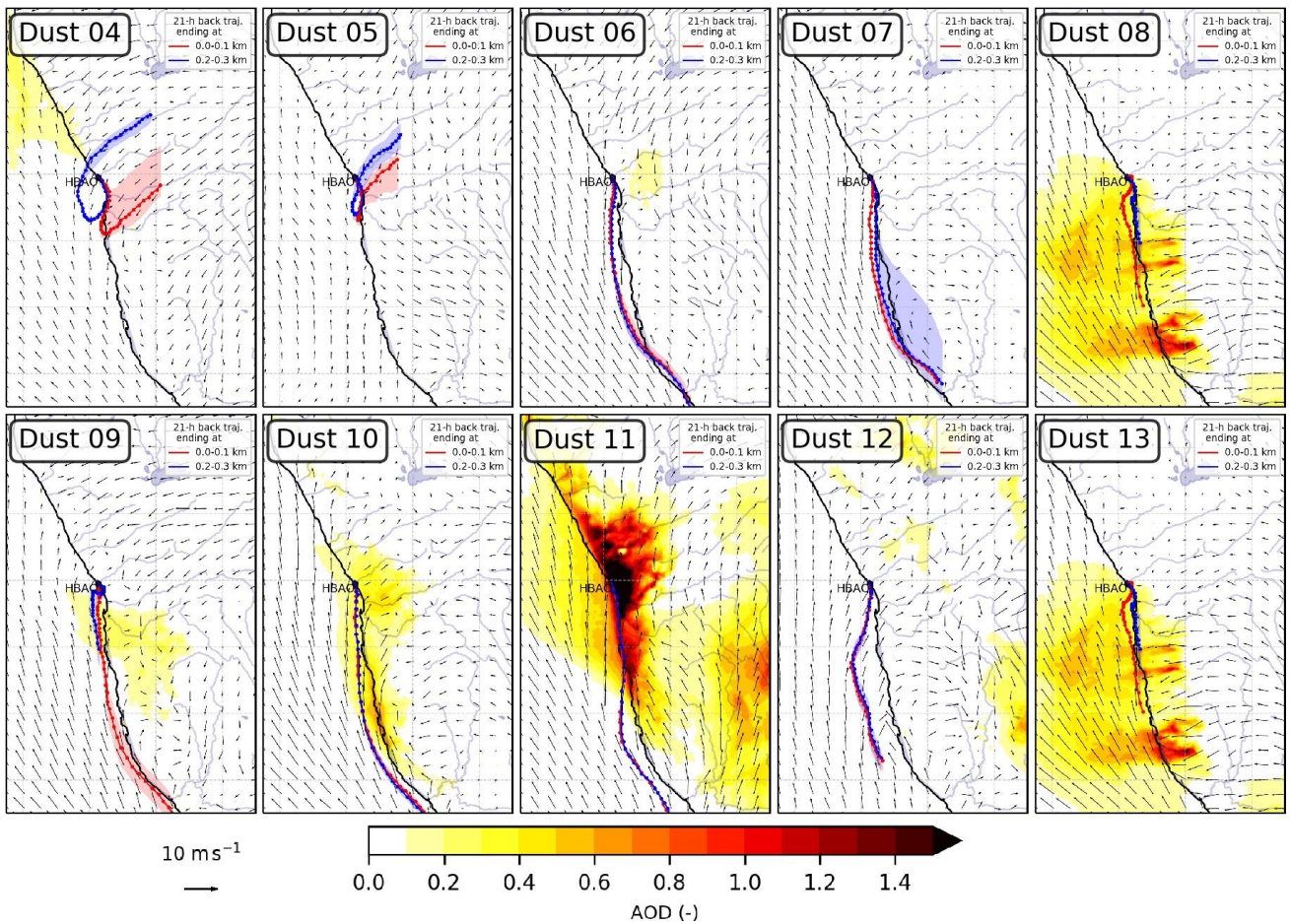



Figure 2.: Maps of dust optical depth (shading) and 10-m wind (vector) overlaid by pathway of 21-hour air mass back trajectories ending in the first 100 m (red line) and between 200 and 300 m (blue line) above HBAO for dust episodes, as calculated by the Meso-NH model (version 5.3). Dots are plotted every hour and shadings around these lines are the interquartile ranges for latitudes.

From July onwards, the active source areas were identified in the southern gravel plains, Namib sand dunes and Kalahari Desert (this former source only for Dust 11 to 13). Air mass transport was southerly and travelled over the sea and along the coastline. It is worth noticing that all the air masses experienced maritime air during their last hours of transport, including the episodes Dust 04 and 05 associated with berg wind conditions, due to the coastal low that develops to the west of HBAO.

The formation of fog events at Henties Bay is also highly seasonal. The frequency of occurrence of fog events is highest during austral winter at the coast, whereas lifted stratus clouds dominate during austral summer, when overall FLC occurrence peaks. The occurrence of fog over Namibia correspond to the advection of low-level clouds which is modulated both by local meteorology along the coastline of Namibia (trade winds) and synoptic-scale radiative processes (Spirig et al., 2019; Andersen et al., 2019; 2020). Henceforth, as shown in Fig. S1, the presence of fog and low clouds correlates with wind directions and aerosol source regions. Overall, three episodes (Dust 04, Dust 05 and Dust 11 in April, May and November, respectively) occurred in fog-free or low-fog conditions. The remaining episodes were characterised by extensive fog and low cloud coverage throughout the study area. The

270 meteorological observations at the nearby Wlotzkasbaken station (Fig. S2) confirm these findings,

271 and show in particular that the relative humidity always exceeded 60 %, and 80 % when fog or low

272 clouds were present (Table 1). As a consequence, the aerosol can be considered deliquescent even

273 in the fog-free conditions. The seasonality is also observed in the average downwelling solar irradi-

274 ance, with the lowest values during July and September, associated with austral winter. Finally, it is

275 interesting to note that the fog-free conditions, associated with the predominance of continental air

276 masses, corresponded to the highest ~~estimated dust mass (~~EDM~~)~~, possibly because of the reduced

277 wet removal during transport and the increase of emission fluxes with the decrease of soil moisture

278 (Kok et al., 2014), but possibly also because of the high wind speed prevailing during these conditions,

279 which in principle, enhances both dust emissions and transport (Table 1).

3.2.   Iron solubility

281 The total and dissolved concentrations, and fractional solubility of Fe, Al and Si, during the dust epi-

282 sodes are reported in Table 2, where they are compared to background conditions. For iron, the

283 average values over the entire sampling period are also shown.

284 **Table 2.** Average and standard deviations of water-soluble (DX), total elemental (TX) mass concentrations and
285 fractional solubility (%SX) for Fe, Al and Si at HBAO measured for the total period and during the dust and
286 background events from April to December 2017. Concentrations values are expressed in ng m^{-3}, while frac-
287 tional solubility is expressed in percent. The numbers of considered samples is presented between the paren-
288 theses.

	Fe			Al		Si	
	All period	Dust	Background	Dust	Background	Dust	Background
DX	28 ± 51 (N=175)	80 ± 84 (N=42)	11 ± 10 (N=131)	322 ± 296 (N=42)	56 ± 46 (N=131)	529 ± 616 (N=42)	78 ± 83 (N=124)
TX	364 ± 482 (N=176)	955 ± 633 (N=42)	177 ± 155 (N=133)	1204 ± 870 (N=42)	284 ± 222 (N=94)	4158 ± 3037 (N=42)	776 ± 674 (N=133)
%SX	7.1 ± 3.6 (N=175)	7.9 ± 4.1 (N=42)	6.8 ± 3.3 (N=130)	27 ± 10 (N=42)	26 ± 11 (N=90)	12 ± 7 (N=42)	11 ± 8 (N=116)

290 The total Fe concentrations varied significantly from one episode to the other, and so did EDM, which

291 was larger than 10 µg m^{-3} for all of them (except Dust 12) and as high as 56 µg m^{-3} during Dust 11

292 event (Table 1). By contrast, the total Fe-to-EDM ratio was virtually constant, with an average of 5.8

293 % (± 0.6 %) for the dust events and 5.6 % (± 1.1%) for the entire dataset. These values are quite

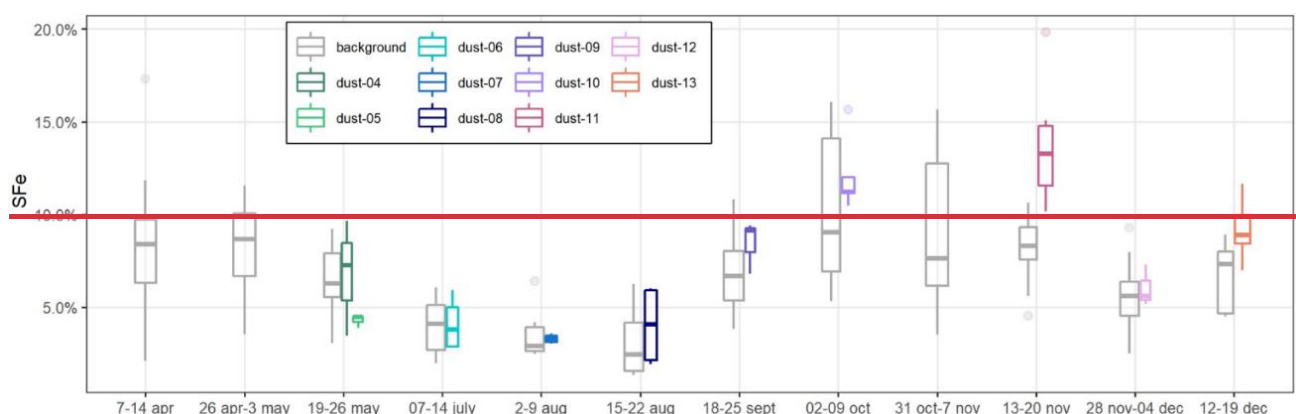
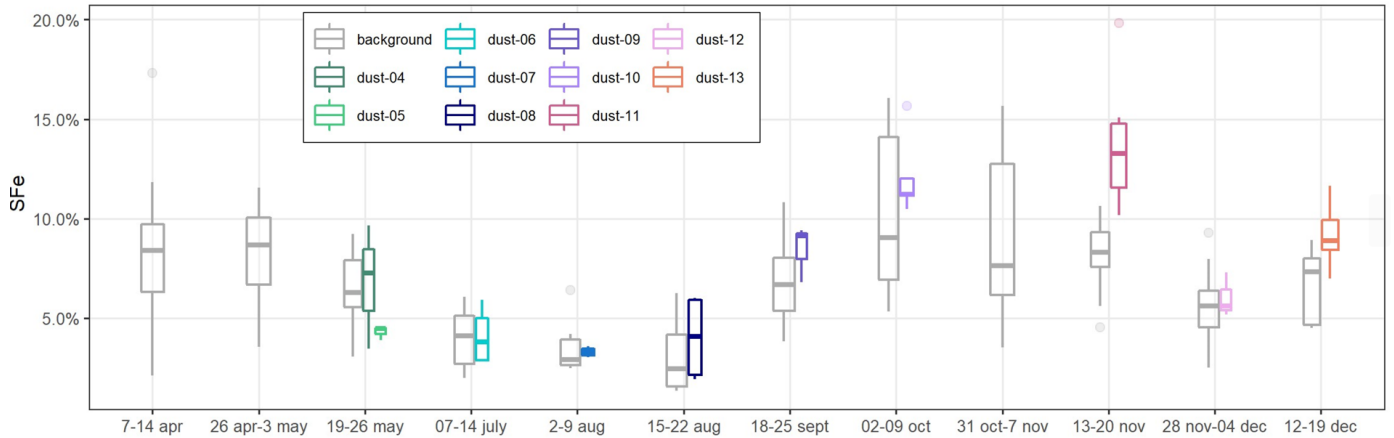
294 superior to usual Fe content recommended in upper continental crust models (3.5% for Taylor and

295 McLennan or 5.04 +/- 0.53 % Rudnick and gao, 2004) and estimated in Saharan dust (4.45% for

296 Guieu et al., 2002; 4.3 to 6.1% for Lafon et al., 2006 or 4.5% for Formenti et al., 2008). Keeping in

297 mind that Fe abundance is estimated, this suggests that the Namibian aerosol dust could be enriched

298 in iron in comparison to upper crust and dust provided by Saharan sources.

The total dissolved concentrations of Fe during the sampling period ranged from 1.5 to 427 ng m^{-3}, with a median and average of 10.5 and 28 ng m^{-3}. During the dust episodes, the average mass concentration of dissolved Fe was 80 ± 84 ng m^{-3}, almost an order of magnitude higher than for background conditions (11 ± 10 ng m^{-3}). The dissolved concentrations in dust periods are higher than those observed in the South Atlantic Ocean for air masses associated with transport from continental southern Africa (Baker et al., 2013; Chance et al., 2015; Baker and Jickells, 2017), which are of the order as those observed at HBAO for background periods. The calculated %SFe ~~fractional solubility of Fe~~ ranged from 1.3 to 19.8 %, with a median and average of 6.7 and 7.1 %. The average %SFe during dust events (7.9 ± 4.1%) was higher, but quite similar than in background conditions (6.8 ± 3.3%). It is interesting to note that Dust 11 event, the most intense recorded event, presents the highest %SFe (between 10.2 and 19.8 % with an average at 13.8 %). Apart from this event, the average fractional solubility seems to be independent of the EDM. Excluding this event, the average solubility of Fe for dust events (6.9 % ± 3.3 %) is equivalent to the one for background samples. The uniformity of %SFe ~~iron solubility~~ values between background and dust periods contrasted with the observations made in regions where the dust influence is sporadic and the origin of Fe is associated to various sources (e.g. Shelley et al., 2018). That is consistent with a main dust source of iron in our samples, as indicated in KL20. For both conditions, the observed range of variability is high and consistent with previous observations over the Southern Atlantic Ocean (2.4-20 %, Baker et al., 2013; 1.3-22 %, Chance et al., 2015), and the Southern Indian Ocean (0.76-27 %, Gao et al., 2013), using acetate buffer leach at pH 4.7 (0.4µm) which can extract 1.4 times more Fe than UPW protocol (Perron et al., 2020). Moreover, the measured %SFe ~~iron fractional solubility~~ is significantly higher than obtained from dissolution experiments, with an identical protocol, of mineral dust aerosol samples collected on filters after laboratory generation from the soils collected in Namibian sources (< 1%; Formenti et al., in preparation, 2024).

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325
326 **Figure 3: Temporal variability of %SFe average for dust and background samples during the different**
327 **periods of sampling. In the box plots, the box indicates the interquartile range, i.e. the 25th and the 75th**
328 **percentile, and the line within the box marks the median. The whiskers indicate the quartiles ±1.5 times**
329 **the interquartile range. Points above and below the whiskers indicate outliers outside the 10th and 90th**
330 **percentile.**

331

332 The temporal variability of %SFe is presented in Fig. 3, where dust and background episodes are

333 shown separately. The temporal variability is similar during dust and background conditions. The

334 highest %SFe occurred during austral spring (October-November), and in particular during episode

335 Dust 11 from 13 to 20 November 2017, when the average %SFe reached 13.8 %. The %SFe was

336 quite similar along the year between dust and background, except between 13-20 November where

337 the iron solubilities during Dust 11 event was very superior to the one of background samples, and to

338 a lesser extent, in September (Dust 09) and December (Dust 13).

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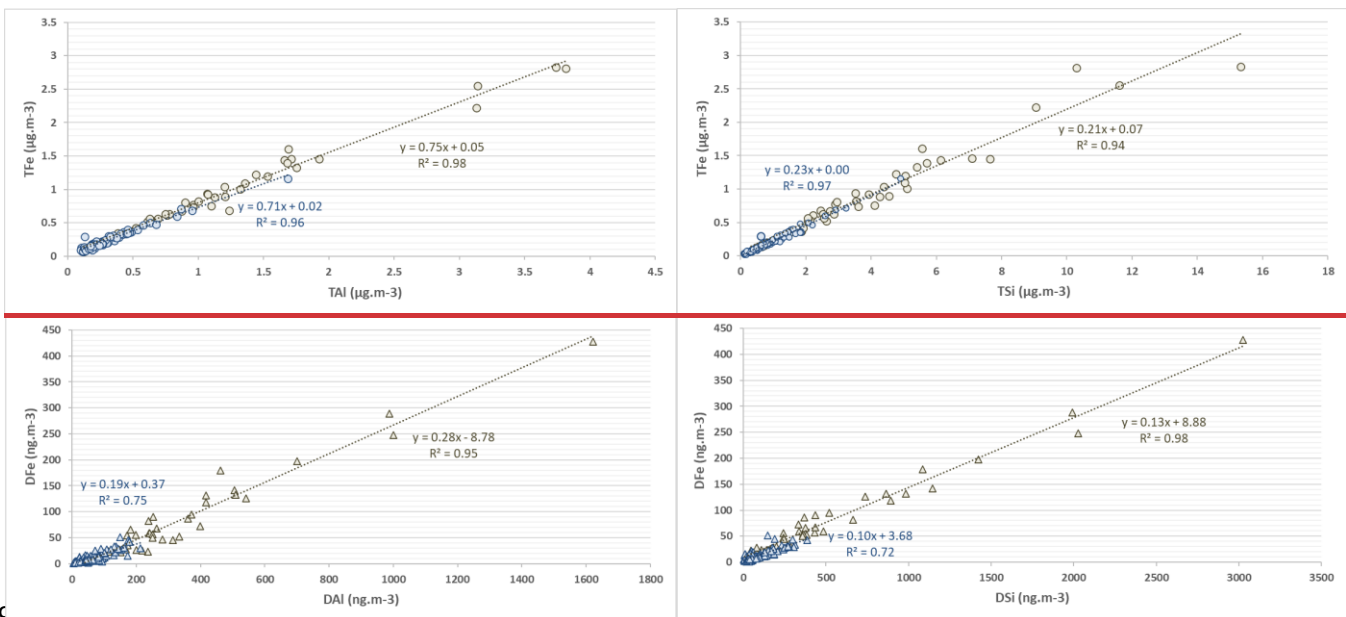
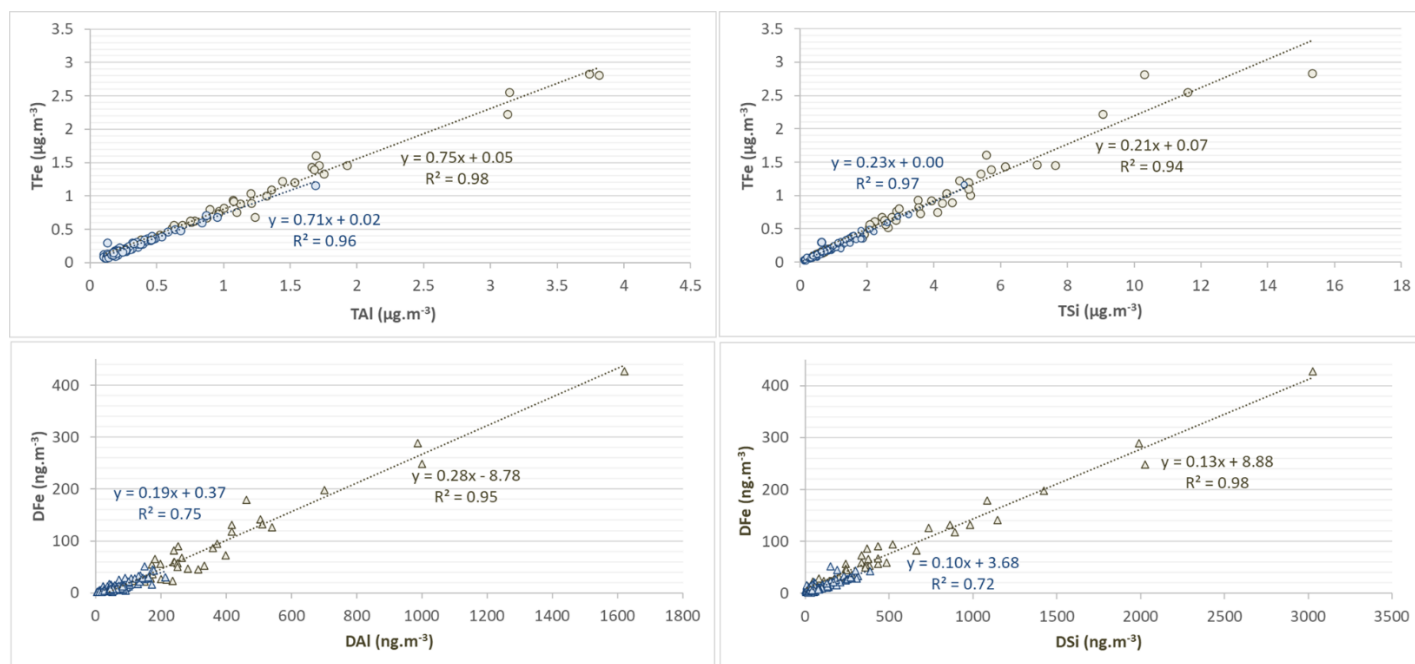

Fig­ure 4­: Scatterplot of TFe with respect to TAI and TSi (top panels) and DFe with respect to DAI and DSi (bottom panels) for dust (sand dots and triangles) and background events (blue dots and triangles). The Pearson coefficient are shown for both.

Fig­ure 4 represents the correlations of Fe with Al and Si, both for the total and the dissolved concen­trations. For both dust and background samples, the total Fe concentration is linearly correlated with total Al (R^2=0.98 and 0.96, slope=0.75 and 0.71, for dust and background conditions respectively) and total Si (R^2=0.94 and 0.97, slope=0.21 and 0.23, respectively). The slopes are consistent with typical Fe/Al and Fe/Si ratios found in desert dust from northern Africa (Formenti et al., 2011; Shelley et al., 2014), confirming the main crustal origin of Fe during all the sampling periods. Likewise, the concentrations of dissolved iron (DFe) show a strong linear correlation with both DAI and DSi, for both for dust and background events (R^2=0.96 and 0.75 with respect to DAI and R^2=0.98 and 0.73 with respect to DSi). The slopes for Al and Si are also comparable (0.19 and 0.28 for DAI and 0.10 and 0.13 for DSi, respectively in dust and in background events). A very strong linear correlation was also observed between DFe and DTi (R^2=0.96 and 0.84; not shown), another unique marker of mineral dust. Significant correlations of soluble concentrations for several elements associated with mineral dust (Fe, Al, Si, Ti) have been previously obtained in remote aerosols over oceanic areas (Baker et al., 2016). Additionally, DFe during dust events correlate very closely with F^- (R^2=0.94, not shown), which has been indicated by KL20 as being emitted in the atmosphere by the wind erosion as well as the labouring of the Namibia soil, rich in fluoride mineral deposits.

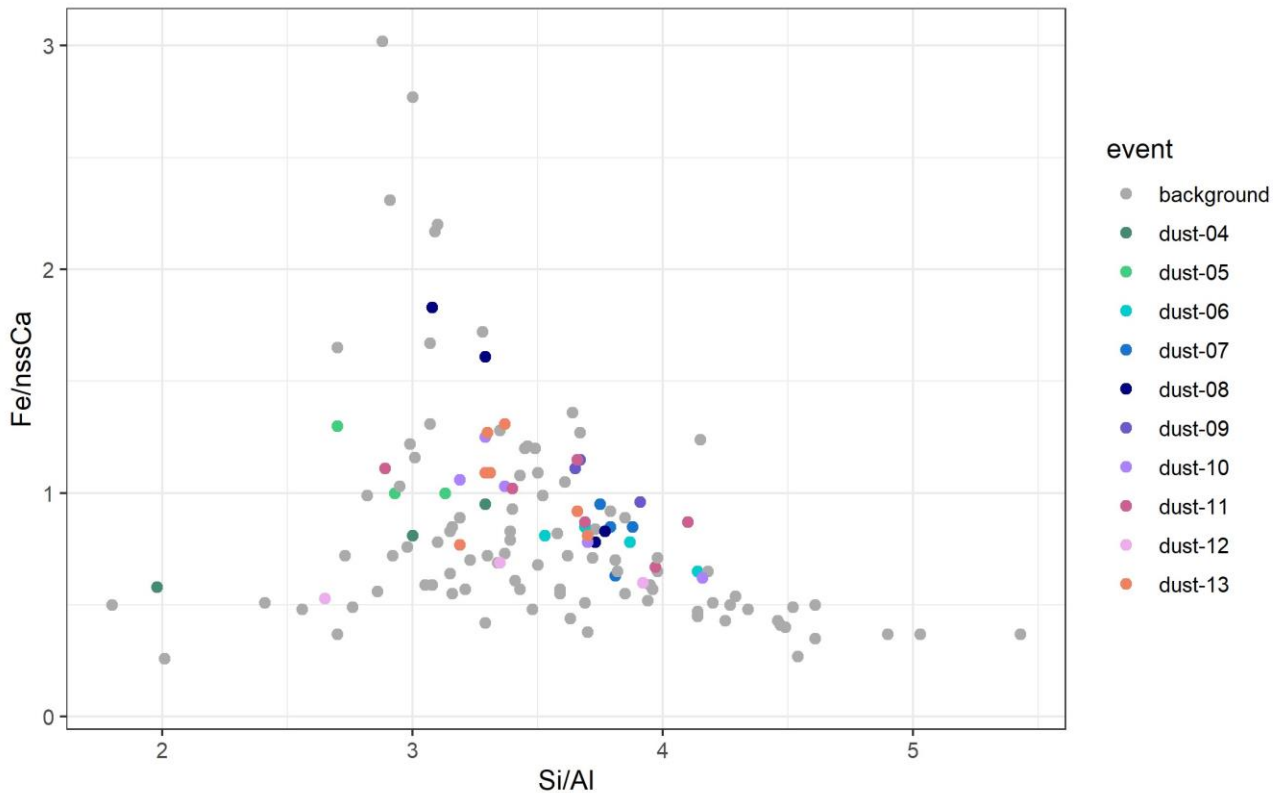
4.   Discussion

Several studies have showe̶d̶n̶ that variations in aerosol Fe solubility could result from the source/composition of the aerosols. As a matter of fact, the Fe solubility has been linked to the iron

mineralogy (Journet et al., 2008) and has been shown being lower for African crustal sources than in continental/anthropogenic sources (Desboeufs et al., 2005; Sholkovitz et al., 2009; Shelley et al., 2018). The iron fractional solubility in mineral dust is also affected by source mixing (Paris et al., 2010; Desboeufs et al., 2005), by (photo)chemical processing with acids or organic ligands during atmospheric transport (Paris et al., 2011, Paris et Desboeufs, 2013; Wozniak et al., 2013; Swan and Ivey, 2021) and by the increase of surface area to volume ratio due to size changes during transport (Baker & Jickells, 2006; Marcotte et al., 2020).

In the following sections, we discuss these possible factors to explain the seasonality and the extended range of variability of the ~~fraction Fe solubility~~%SFe in HBAO samples. The possible increase of surface area to volume ratio during transport (Baker and Jickells, 2006; Marcotte et al., 2020) will not be discussed because of lack of appropriate observations of the size distribution. Because of the similar transport time suggested by back trajectories (Fig. 2), it is likely that particle size distribution would be similar from one event to the other.

4.1. Influence of dust composition

Close to the dust source, iron solubility could be mainly conditioned by the mineralogical composition of dust (Journet et al., 2008, Formenti et al., 2014). Considering that soluble Fe-bearing aerosols were issued from mineral dust for all the samples, the seasonality of dust emission sources (see 3.1) could be a factor explaining the seasonality of %SFe (and other elements associated to mineral dust). Fig. ure 5 shows the scatter plot of the elemental mass ratio of Fe/nss-Ca^{2+} and Si/Al, previously used for northern Africa dust to distinguish aerosol dust from source areas enriched in clays or iron oxides to soils rich in quartz or carbonates (Formenti et al., 2014). Specific to Namibia, because of the strong link between nss-Ca^{2+} and fluorine, the Fe/nss-Ca^{2+} ratio may also to distinguish dust influenced by fluorspar mining.

388

389 ‑Fig**ure** 5**.:** Scatterplot of Fe/nss-Ca²⁺ and Si/Al mass ratios for the samples collected at HBAO in period
390 **May-December 2017. Values obtained for samples collected during the dust events are represented as**
391 **colored dots. Values for samples collected outside those events (background) are represented as grey**
392 **dots.**

393

394 Figure 5 indicates that the range of variability of both Fe/nss-Ca²⁺ and Si/Al ratios is small when con-

395 sidering dust events only. The elemental ratios of samples collected during the background periods

396 are rather similar to dust events during a same sampling period, except for Si/Al for the period be-

397 tween 19-26 May and for Fe/nss-Ca²⁺ for the samples of 18-25 September, when significant differ-

398 ences, not really explicable and not inducing a significant difference in the %SFe values are observed

399 (Fig. S3).

400 The values for ambient dust measured at HBAO are consistent with those of the previous field obser-

401 vations in Namibia (Annegarn et al., 1983; Eltayeb et al., 1993), but also with values reported by

402 Caponi et al. (2017) for laboratory-aerosolised dust from two soils collected on the Namibian gravel

403 plains. This is in agreement of the indications of the emission maps (Fig. 2), showing significant emis-

404 sions in the gravel plains. The absence of seasonal cycle in the elemental composition illustrated in

405 Fig ure S3 suggests that the seasonal change from northern to southern sources does not induce a

406 change in the composition of the aerosol dust sampled at HBAO, which is consistent with the fact that

407 the northern and the southern gravel plains of Namibia have similar mineralogy (Heine and Vökel,

408 2010). This suggests that the mineralogical composition of mineral dust should not be a discriminating

409 factor explaining the seasonality of the iron solubility observed at HBAO.

4.2. Evidence of processing by marine biogenic emissions

The atmospheric (in-cloud) processing associated with secondary aerosol production may increase the fractional solubility of Fe during transport (Takahashi et al., 2011; Rodríguez et al., 2021). This has also been shown for Al and Ti (Baker et al., 2020). The chemical processing could include both acidic and ligand-promoted dissolution (Desboeufs et al., 2001, Longo et al., 2016, Tao et al., 2019). Oxalic acid has previously been used as a proxy for organic ligand–mediated iron dissolution processes because it is the most abundant species in the atmosphere and is the most effective ligand in promoting iron dissolution (Baker et al., 2020; Hamilton et al., 2021). However, several secondary compounds, such as carboxylate ligands and marine secondary products derived from ~~dimethyl sulfide (~~DMS~~)~~ oxidation, have been identified as playing a role in increasing the soluble fraction of iron from mineral aerosols (Johansen and Key, 2006; Paris et al., 2011; Paris and Desboeufs, 2013; Wozniak et al., 2013 and 2015). The increase of ligands-promoted dissolution is attributed to photochemical reduction of Fe(III) in Fe (II) (Siefert et al., 1994; Johansen and Key, 2006).

To investigate these aspects, the mass concentrations of the ionic compounds (oxalate, formate, MSA, NO_3^-, NH_4^+ and $nss-SO_4^{2-}$) implied in the secondary aerosol production, measured at HBAO during dust and background periods are reported in Table 3.

Table 3. Average and standard deviations of mass concentrations of water-soluble ions measured at HBAO during dust and background events from May to December 2017. Concentrations are expressed in ng m^{-3}. The number of samples pertaining to each occurrence is indicated in brackets.

	Dust	Background
$nss-SO_4^{2-}$	1795 ± 762 (N = 42)	1366 ± 505 (N=132)
Oxalate	155 ± 53 (N = 42)	127 ± 35 (N = 132)
Formate	18 ± 6 (N = 40)	16 ± 9 (N = 105)
MSA	64 ± 37 (N=36)	56 ± 36 (N=114)
NO_3^-	205 ± 79 (N=42)	200 ± 138 (N=132)
NH_4^+	192 ± 71 (N=42)	207 ± 98 (N=132)

Oxalate was the most abundant organic compound, followed by MSA, a secondary product of DMS oxidation and a unique particulate tracer of the primary marine biogenic activity (Andreae et al., 1995). On average, organic compounds were equally concentrated in dust and background events. Amongst inorganic species, $nss-SO_4^{2-}$ was the most concentrated compound, with higher values during the dust events than during the background period.

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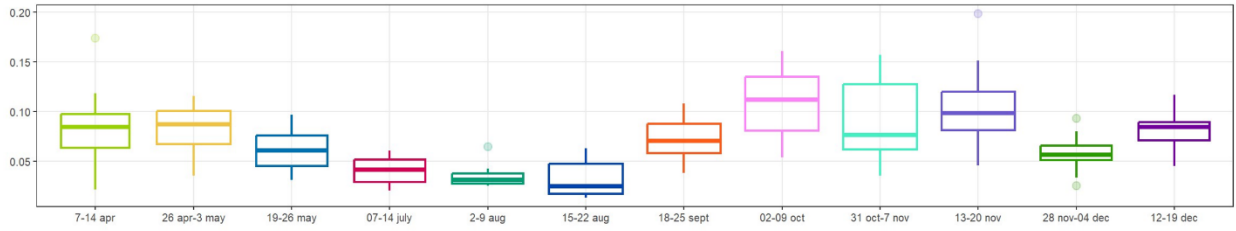
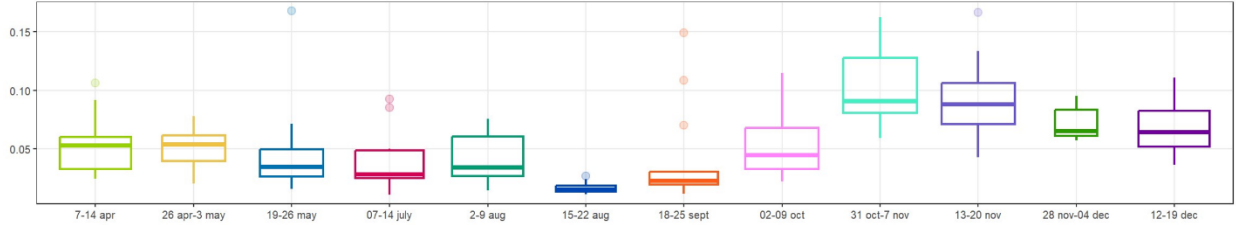
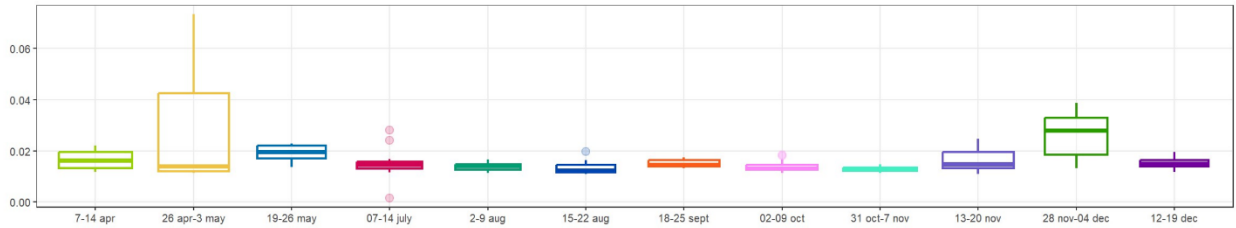
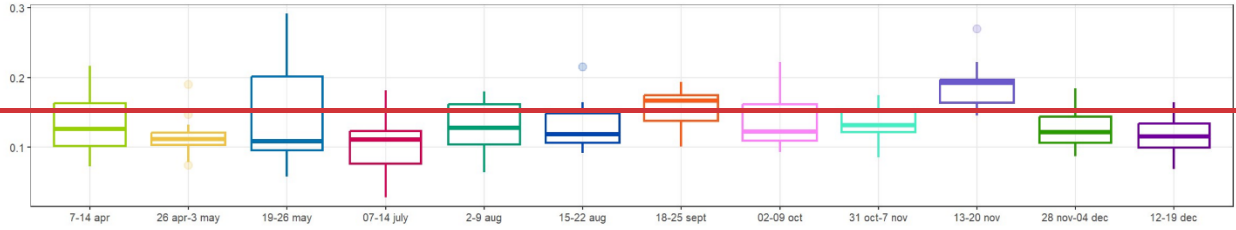
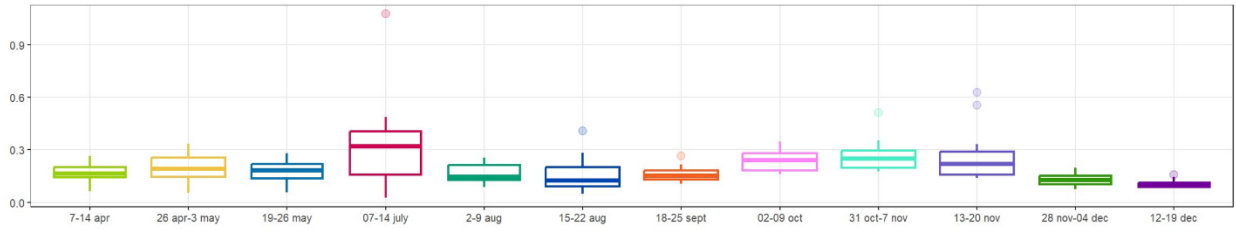
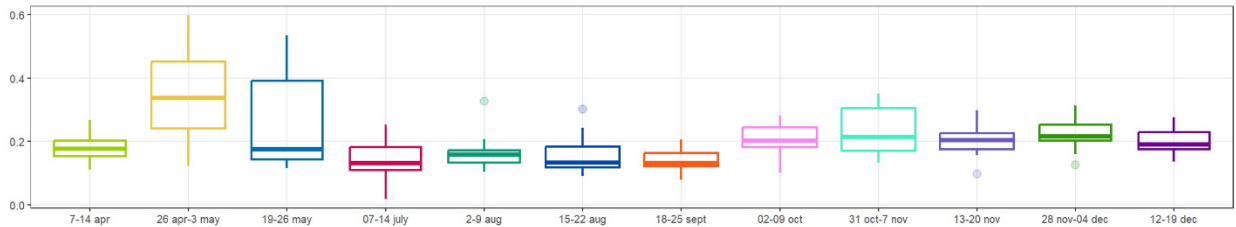
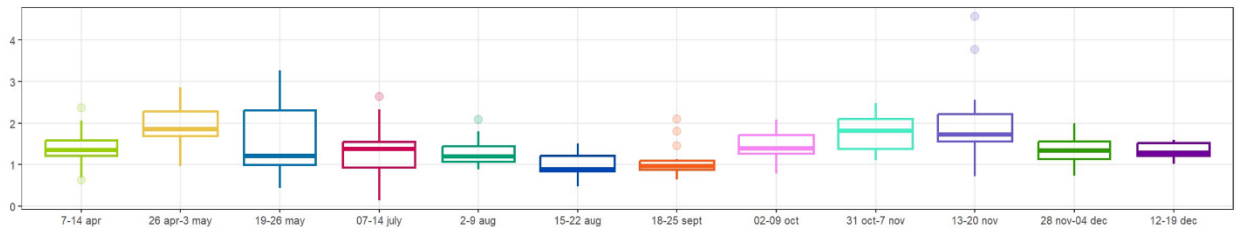
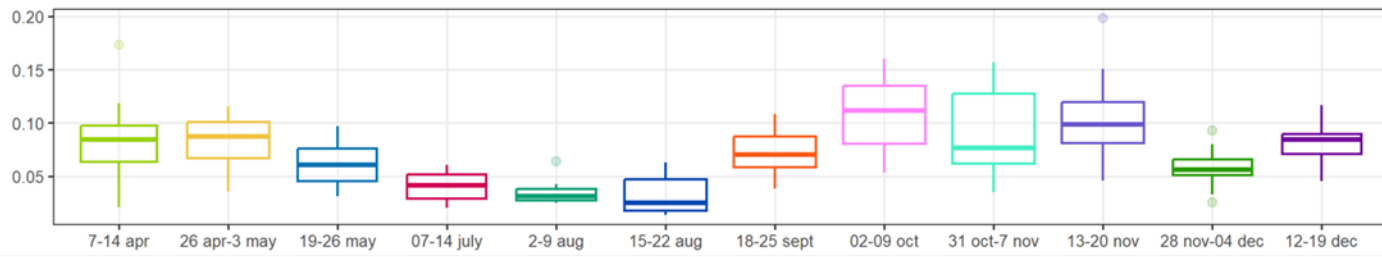
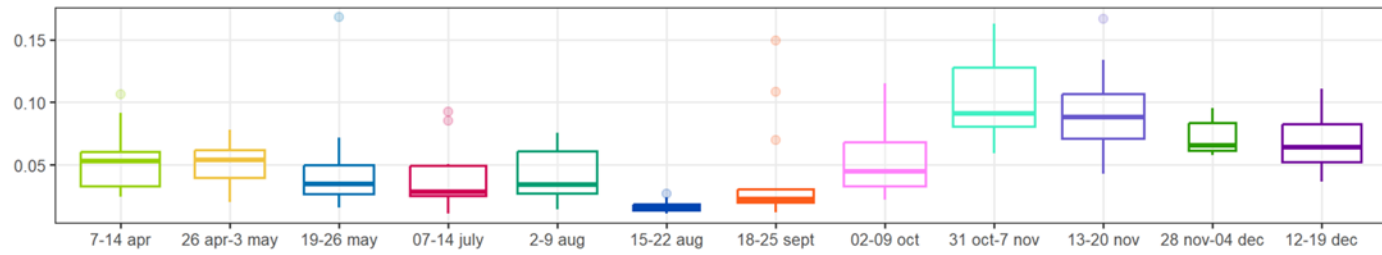
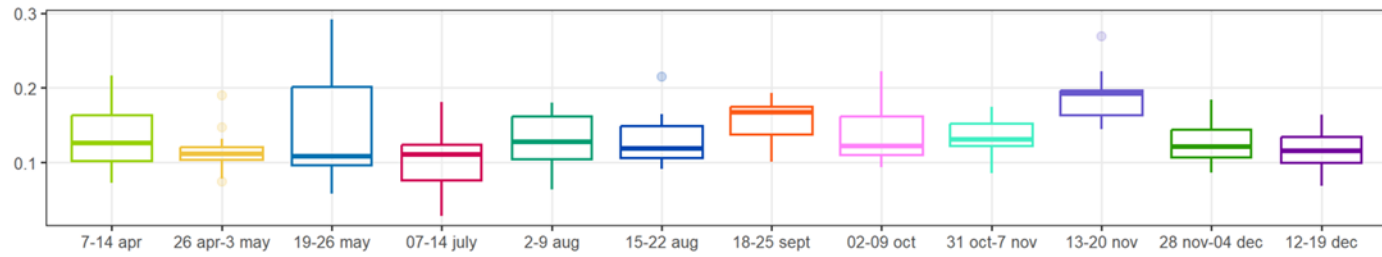
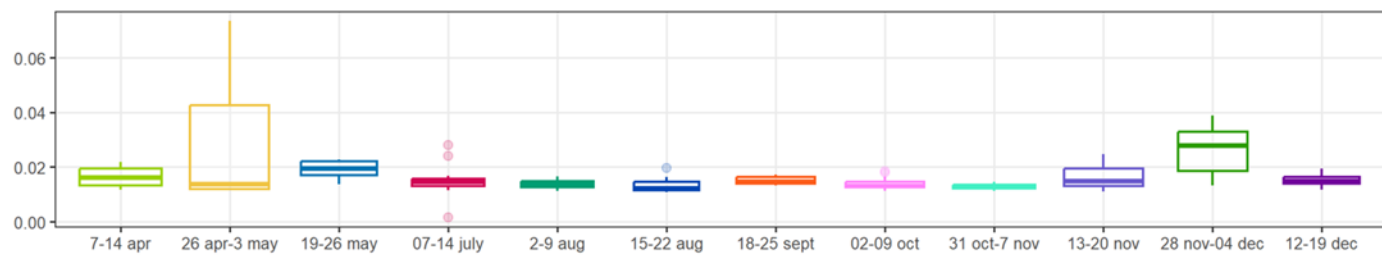
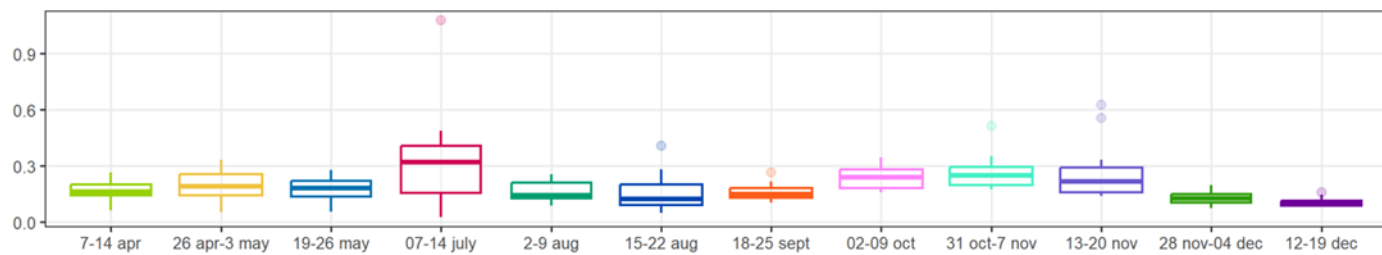
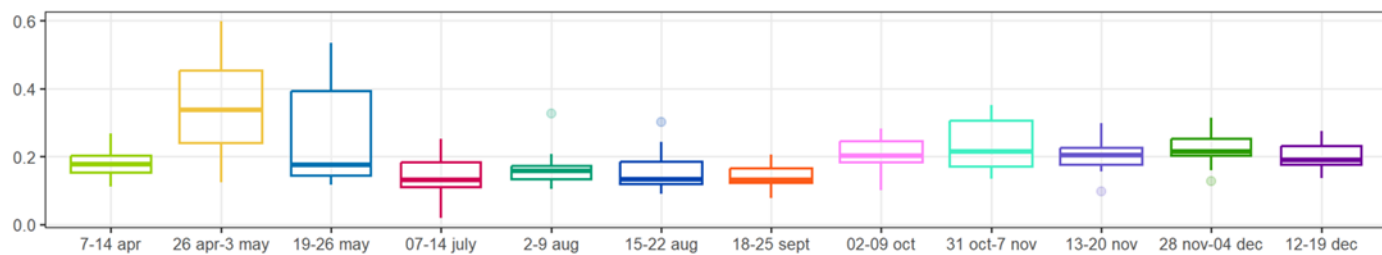
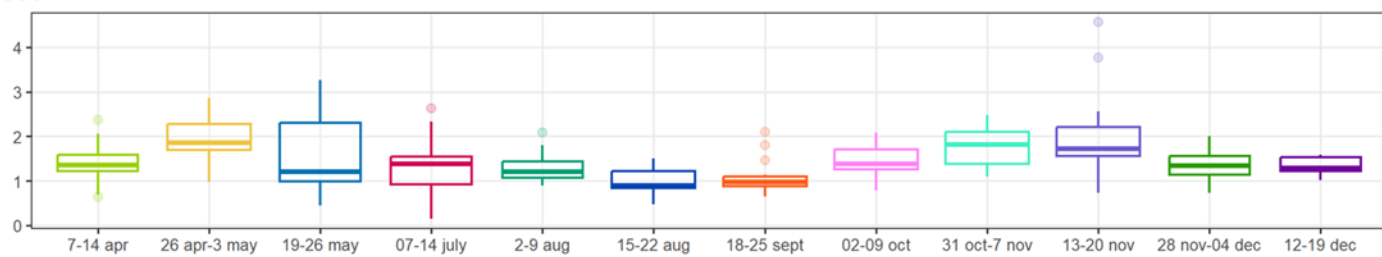
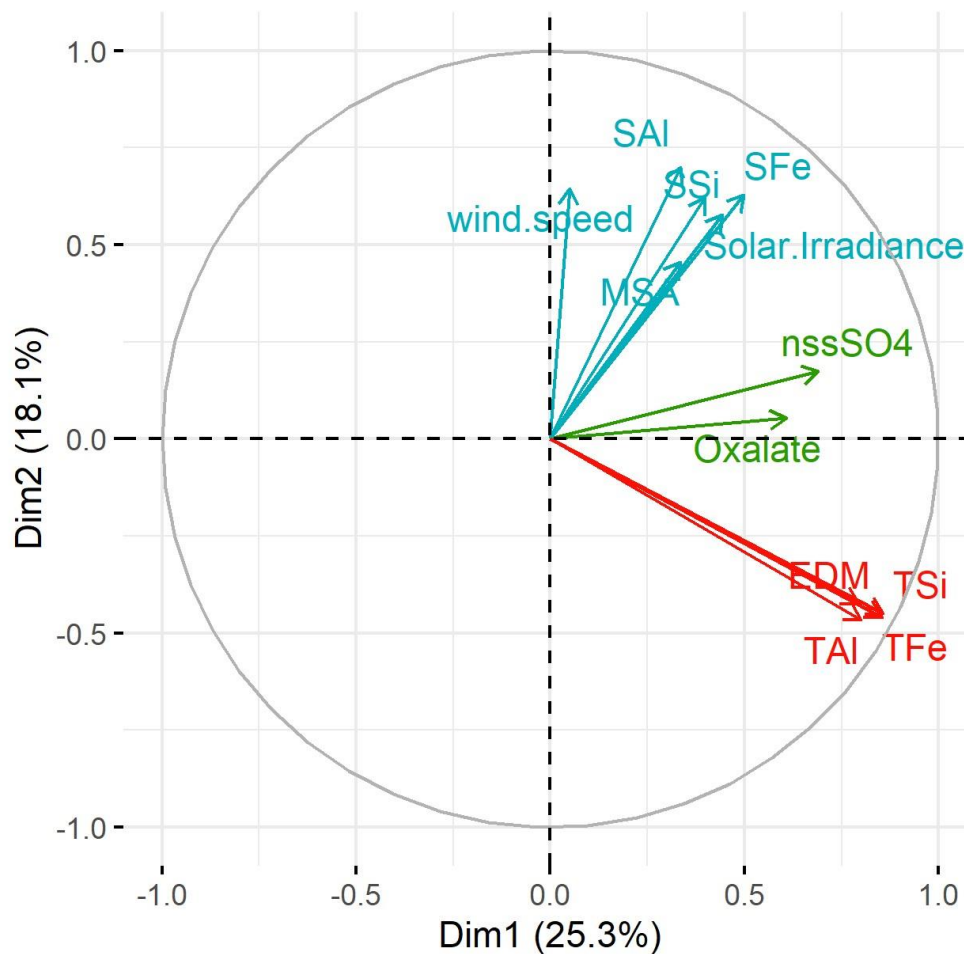


Fig.ure 6.: Box-plots of the averages of %SFe and secondary organic and inorganic compounds mass concentrations (μg m⁻³) for the sampling periods including all the samples (dust + background). Boxes and whiskers as in Fig. 23.

Their detailed time series of secondary compounds are shown in Fig. 6, where it isthey are compared to that of the %SFeiron fractional solubility. There is no clear seasonal cycle for any of the ionic compounds, with the exception of MSA, which shows a similar time variability than %SFe. MSA concentrations were lowest between May and August (average 38.0 ± 28.0 ng m⁻³), while higher concentrations were measured from September to December (72.7 ± 38.1 ng m⁻³). These differences are also observed for the dust cases only. The average MSA concentration was 40.6 ± 23.4 ng m⁻³ for Dust 04 to Dust 08 episodes. It increased to 77.7 ± 35.3 ng m⁻³, almost a factor of 2 between episodes Dust 09 and Dust 13. The mass concentrations and the seasonal cycle of MSA are related with the proximity of the strong coastal upwelling by the Benguela current (Formenti et al., 2019; KL20). The maximum concentration of MSA (106.2 ng m⁻³) was measured during episode Dust 11, which is also the time of the highest SFe% observation. This episode was also characterised by the highest oxalate, nss-SO$_4^{2-}$ and NO$_3^-$ concentrations. However, no clear correlation between the %SFe and the secondary compounds concentrations can be found in our data (Fig. S4). In order to statistically explore the potential links between %SFe and various these concentrationsparameters, Fig.ure 7 shows the correspondence plot between total Fe, Al and Si, and their respective fractional solubility, the measured secondary compounds and the meteorological conditions during sampling obtained from Principal Component Analysis (PCA) for all the samples. The variables correlated in time are grouped together (the closer they are in the circle, the stronger the correlation) whereas the variables which are anti-correlated are situated on the opposite side of the plot origin.

463 **Figure 7.: PCA analysis performed from the database including %SX,TX, EDM, secondary ions concentrations and**
464 **meteorological parameters. The colour of variables by groups is defined by a clustering algorithm, tending to find**
465 **clusters of comparable spatial extent. Each colour corresponds to a cluster of parameters which evolve in the same**
466 **way. Formate, nitrate, ammonium, acetate, humidity, and wind speed are not visible in the plot showing that they**
467 **are not significantly correlated with the other parameters (i.e. their squared cosine < 0.4).**

468

469 The PCA plot (Fig. 7) emphasizes 3 groups of dependent parameters: 1. a high correlation between

470 total Fe, Al and Si concentrations and dust loading (EDM), as previously identified in Table 1 and

471 Fig. 3, 2. a relation between oxalate and nss-SO_4^{2-} concentrations, suggesting a common chemical

472 process of formation, and 3.the dependence between %SFe (%SAl and %SSi), the MSA concentra-

473 tions, the solar irradiance and to a lesser extent with the wind speed. While it is expected that the

474 emission of mineral dust occurs when the wind speed is high, the correlation of %SFe with wind speed

475 is rather surprising since the %SFe is independent of the dust load (Fig. 2 and 7). Fig.ures S4 in the

476 supplementary material present the plots between %SFe, MSA concentrations, solar irradiance and

477 wind speed for background and dust events. The correlation between the wind speed and the MSA

478 concentrations (Fig. S4) is consistent with Andreae et al. (1995), who demonstrated how, in this area

479 due to persistent phytoplankton bloom, the atmospheric concentrations of ~~dimethylsulphide~~ (DMS),

480 the gaseous precursors of MSA, depend on the sea-to-air flux, in turn is determined by the concen-
481 trations in the ocean water and the surface wind speed.

482 As previously mentioned, Johansen and Key (2006) showed an increase of dissolution of ferrihydrite,
483 a proxy of iron(oxy)hydroxide found in desert mineral dust, by photolysis of the Fe(III)-MSIA (methane
484 sulfinic acid) complex, producing MSA and soluble Fe. Zhuang et al. (1992) proposed an increase of
485 iron dissolution by the acidification of aerosol particles associated with ~~dimethylsulphide~~ (DMS~~)~~ oxi-
486 dation. Here, the link between the %SFe ~~Fe fractional solubility~~, solar irradiance and MSA is in agree-
487 ment with the photo-reduction dissolution of Fe by MSA condensation on Fe-bearing dust. Thus, we
488 attribute the %SFe ~~iron fractional solubility~~ seasonality observed at HBAO both to solar irradiance and
489 MSA temporal evolution via this process. It is interesting to note that due to the high correlation be-
490 tween %SFe and %SAl and %SSi, the photochemical processes could also impact the solubility of all
491 element-bearing dust.

4.3. Link to other sources of iron and oxalate

493 Formenti et al. (2018) have shown that in the Austral winter, when the synoptic circulation is domi-
494 nantly anti-cyclonic, air masses laden with light-absorbing aerosols either from ship pollution or bio-
495 mass burning can be transported to HBAO (Formenti et al., 2018). However, the lowest Fe solubility
496 (< 5%) was measured in July and August 2017, and no correlation between the %SFe and the percent
497 mass fraction of iron from sources other than dust can be found in our data (not shown).

498 The mass apportionment of iron reported by KL20 indicates that, during the dust events and the
499 background periods, respectively, 7% and 29% of the mass of total elemental Fe was not associated
500 to mineral dust, but rather to a factor indicated as "ammonium-neutralised component", mostly char-
501 acterised by secondary species, and non-sea-salt potassium (nss-K^+). The PMF (Positive Matrix Fac-
502 torisation) analysis indicated that the "ammonium-neutralised component" was associated to photo-
503 oxidation of marine biogenic emission but also episodically to biomass burning. This component in-
504 cludes oxalate, the most concentrated organic species at HBAO, and the strongest of the organic
505 ligands promoting the photo-reduction of iron in mineral dust, henceforth the increase of its fractional
506 solubility (Paris and Desboeufs, 2013). Surprisingly, excepted individual cases (Dust 13), our analysis
507 does not show this strong link (Fig. 7), and indeed, contrary to the SFe%, the oxalate concentrations
508 measured at HBAO was practically constant with time (in average 0.14+/- 0.04 $\mu g.m^{-3}$). The possible
509 pathways of oxalate formation in this complex atmosphere are numerous through the year, from nat-
510 ural and anthropogenic sources (marine, heavy-oil combustion, biomass burning) and in-cloud and
511 photo-oxidative processes (Baboukas et al., 2000; Myriokefalitakis et al., 2011).

5.   Concluding remarks

513   For the first time, the fractional solubility of Fe in airborne atmospheric aerosols smaller than 10 μm

514   in diameter is investigated along the west coast of Namibia, in southern Africa, a critical region for the

515   global climate.

516   Ten intense episodes of transport of mineral dust from aeolian erosion were identified from the anal-

517   ysis of aerosol samples collected between May and December 2017 at the Henties Bay Aerosol Ob-

518   servatory (HBAO). Based on modelling and measurements, source regions were identified both in the

519   northern and southern gravel plans. Our data do not provide any evidence of the possible contribution

520   of dust from coastal riverbeds, which are considered to be frequent sources of atmospheric dust and

521   soluble iron in the region (Vickery et al., 2013; Von Holdt et al., 2017; Dansie et al., 2017a; 2017b).

522   The total iron represents, on average, 5.8 % (± 0.6 %) of the total dust mass, and that the average

523   water-soluble Fe fractional solubility is 6.9 % (± 3.3 %). These values should be useful to atmospheric

524   models estimating the dust-borne input of soluble Fe from the gravel plains in Namibia to the sur-

525   rounding oceans.

526   The seasonal increase of the %SFe ~~iron fractional solubility~~ is associated to that of the concentrations

527   of MSA and correlated to meteorological parameters such as the wind speed and the surface solar

528   irradiance. Our observations support the role of photo-chemical processes in the dissolution of Fe in

529   our samples, and suggest that the oxidation of the marine biogenic emissions from the northern Ben-

530   guela upwelling, favoured under high wind speed conditions, could play a significant role in increasing

531   the solubility of elemental iron in mineral dust aerosols over coastal Namibia. This is in agreement

532   with the mechanism described by Zhuang et al. (1992), who proposed an increase of iron dissolution

533   by the acidification of aerosol particles associated with DMS oxidation, and Johansen and Key (2006),

534   who showed an increase of dissolution of ferrihydrite, a proxy of iron(oxy)hydroxide found in desert

535   mineral dust, by photolysis of the Fe(III)-MSIA (methane sulfinic acid) complex, producing MSA and

536   soluble Fe. It is interesting to note that due to the high correlation between %SFe, ~~and~~ %SAl and

537   %SSi (and %STi), the same photochemical processes could also impact the solubility of all element-

538   bearing dust. The possible mechanism suggested by this paper could be responsible for initiating a

539   feedback loop whereby the input of dust of increased trace and major elements solubility would result

540   in stronger marine biogenic emissions to the atmosphere. This possible mechanism could increase

541   the iron solubility in mineral dust, maybe also initiating a feedback loop whereby the input of dust of

542   increased solubility would result in stronger marine biogenic emissions to the atmosphere, including

543   Volatile Organic Compounds ~~(VOCs)~~, in particular butene, massively emitted by the organisms in the

544   coastal marine foam (Giorio et al., 2022).This complex and dynamic environment where the interplay

545   between the input of atmospheric iron from transported dust and the marine biogenic emissions from

546   the Benguela oceanic upwelling system should be further addressed by future research.

547

Data availability. ~~Original and analysed data are available at the AERIS (https://aeroclo.aeris-data.fr/project/, last accessed 20/07/2023)~~Atmospheric concentrations of total and dissolved elements and water-soluble ions measured over coastal Namibia in 2017 are available in the Easy Data (Formenti et al., 2023, https://doi.org/10.57932/2ac79cd1-282a-4004-87d5-38f0ebcaf40c). The statistical FactoMineR package is available in R (R version 4.1.2, 2021; http://factominer.free.fr/index_fr.html, last accessed 20/07/2023). Meteorological data from the Wlotzkasbaken station (22.31°S, 14.45°E, 73 m asl) are part of the Southern African Science Service Centre for Climate Change and Adaptive Land Management (SASSCAL) ObservationNet (https://www.sasscal.org/; last accessed 14/04/2023).

Author contributions. PF, DK, SJP, AN, MC, AF and SC prepared and performed the filter sampling. RT, KD, PF, SC, ST,and CMB performed the XRF, IC and ICP analysis of the collected samples. BL performed the field implementation. KS and SF performed the model calculations of dust emission fluxes. JPC performed the model calculations of air mass back-trajectories. HA and JC provided with the satellite retrieval of fog and low clouds. PF, KD, RT and SJP analysed and interpreted the dataset. PF and KD wrote the paper with contributions from RT and SJP, and the remaining authors. PF and SJP provided funding. PF coordinated the research activity and supervised its planning and execution.

Competing interests. PF is guest editor for the ACP Special Issue "New observations and related modelling studies of the aerosol–cloud–climate system in the Southeast Atlantic and southern Africa regions". The remaining authors declare that they have no conflicts of interests.

Special issue statement. This article is part of the special issue "New observations and related modelling studies of the aerosol–cloud–climate system in the Southeast Atlantic and southern Africa regions (ACP/AMT inter-journal SI)". It is not associated with a conference.

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590

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