1 Emissions from potential Patagonian dust sources and

- associated biological response in the Atlantic sector of the
 Southern Ocean
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15 Abstract

The effect of Patagonian dust over primary producers in the Southern Ocean has long been 16 17 disputed. Here we present new remote sensing evidence in favour of dust mediated biological 18 response and postulate a hypothesis to explain the spatial relation observed. A new remote 19 sensing definition of dust source areas based on the Normalized Difference Vegetation Index 20 (NDVI) and Absorbing Aerosol Index (AAI) correlation is presented and interannual 21 variation in AAI is evaluated within the source regions as a proxy for dust activity. 22 Correlation of this data with annual chlorophyll concentration, phytoplankton biomass, and 23 diatom dominance reveals a spatially coherent latitudinal band of positive correlation 24 concentrated between the Polar Front and the Subtropical Front. This pattern is restricted to western areas in the biomass correlation and extends toward Africa for the chlorophyll and 25 26 diatom correlation. This region is equivalent to the area of the Subantarctic Mode Water formation, characterized by a ratio $Si:N \ll 1$ in late summer, an unfavourable condition for 27 28 diatom development, especially under iron limitation. Therefore, due to Si-Fe co-limitation, 29 the positive correlation could be the consequence of an enhanced sensibility of this area to

external iron addition for diatom growth. For the Argentinean shelf-break, is not clear
 whether direct dust input and/or wind stress driving water masses upwelling could be
 responsible for the positive correlation.

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5 **1** Introduction

6 The last three decades saw an emerging discussion on the role of micronutrients, particularly 7 iron, as a limiting factor for marine biological production (e.g., Martin and Fitzwater, 1988; de Baar et al., 1995; Boyd et al., 2007; Westberry et al., 2013). Iron (Fe) is a co-factor of 8 9 metaloenzimes on major biochemical pathways, from respiratory electron transfer reactions to 10 chlorophyll synthesis (Geider and La Roche, 1994; Morel et al., 2003). Although it is the 11 fourth most abundant element on the Earth's crust (Rudnick and Gao, 2003), at the current oxidizing conditions of the atmosphere and upper ocean, it is present primarily as Fe^{+3} , which 12 is largely insoluble in seawater (Liu and Millero, 2002). As a result, the large amounts of 13 14 eroded iron added to coastal waters are removed by physical, chemical and biological 15 processes before reaching the open ocean.

In the Southern Ocean, where Ekman divergence drives strong upwelling of deep water masses rich in dissolved macronutrients (Orsi et al., 1995; Sigman et al., 1999), observational (e.g., Pollard et al., 2009) and experimental evidences (e.g., Martin et al., 2013) have shown that additions of iron alone can modify ecosystem functioning (Moore et al., 2013), enhancing primary production, with potential consequences to the carbon cycle (Smetacek et al., 2012).

21 Natural direct input of iron to these waters occurs through diffusion and mixing of dissolved 22 iron from seabed and direct aeolian deposition of mineral dust into the surface (e.g., Moore 23 and Braucher, 2008). In the Atlantic sector of the Southern Ocean, a region over the influence 24 of Patagonia (Li et al., 2008), models suggest that dust can be as important as oceanic sources even at the expected modern low dust flux (Fung et al., 2000; Moore and Braucher, 2008). 25 26 This is a unique region of the Southern Ocean in which geological evidence points to a 27 coupled variation of dust flux and biological production over the last 1 My (Maher et al., 28 2010).

29 However, evidences are not consistent and the relative contribution of these sources, and their

30 impact on the ecosystem, has been a matter of intense debate over the last decade (Fung et al.,

31 2000; Erickson et al., 2003; Cassar et al., 2007; Meskhidze et al., 2007; Johnson et al., 2011).

Much of this debate is based on modelling studies, which, despite their relevance, are subject to large errors. For example, dust iron solubility is a parameter notoriously difficult to simulate, as it is dependent on its mineralogy (Journet et al., 2008), physicochemical reactions during transport (e.g., Baker and Croot, 2010), plankton diversity (e.g., Rubin et al., 2011) and chemical condition of the ocean (Bressac and Guieu, 2013). But even the dust emission and flux are largely uncertain, and modelled South American dust emission can vary by 10³ Tg yr⁻¹ between models (Huneeus et al., 2011).

8 Here we present a remote sensing approach to study if and where the interannual variation on 9 dust emission from Patagonia <u>may</u> exerts observable signal on the phytoplankton in the 10 Atlantic sector of the Southern Ocean.

11 2 Methods

12 **2.1** Source areas and dust activity

13 Dust and other iron containing bearing aerosols show a characteristic absorption in the blue-14 ultraviolet (UV) region of the electromagnetic radiation, which increases toward more 15 energetic wavelengths (Patterson, 1981). Therefore, the difference in the observed and 16 modelled clear sky spectral contrast in two UV bands can be used efficiently for their 17 identification (Herman et al., 1997). The resulting parameter, the Absorbing Aerosol Index 18 (AAI), can be is regarded as a qualitative semi-quantitative proxy (Torres et al., 2002) 19 (Chiapello and Moulin, 2002), since beside the dust load, the microphysical properties of the 20 aerosol and the altitude of the aerosol layer also have significant influence on its absolute 21 value (Torres et al., 1998; De Graaf et al., 2005). However, for temporal monitoring over 22 source regions, microphysical properties and mean altitude should present a much lower 23 variability than dust optical thickness, allowing a quantitative use of the index. Such approach 24 have already has been verified (e.g., Chiapello et al., 1999; Hsu et al., 1999; Deroubaix et al., 25 2013) and employed with success in other studies (e.g., Chiapello et al., 1999 Chiapello and Moulin, 2002). 26

We use the scientific AAI product (version 5.1) from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) sensor on-board the Envisat platform (Tilstra et al., 2012), to calculate interannual variation from 2003 to 2010. We exclude 2011 from the calculations due to the intense Puyehue (Chile) eruption initiated in June, which covered a vast area with thick ash deposits (Gaitán et al., 2011), prone to
 remobilization by wind erosion (Haller and Frumento, 2012).

3 To define the dust source areas within meridional South America we relied on the negative 4 correlation of the AAI with a vegetation proxy, employed as an integrative parameter of two 5 time varying surface properties related to dust emission (Jobbágy et al., 2002; Cropp et al., 6 2013): (i) the soil moisture content, which influences particle cohesion; and (ii) the abundance 7 and structure of vegetation, which influences the transmittance of the kinetic energy from the 8 wind to the surface (Tegen and Fung, 1994; Mahowald et al., 2005). Together, these 9 parameters regulate the threshold wind velocity needed to initiate the dust emission over a 10 specified region. An analysis highlighting areas where AAI increase is related to vegetation 11 decrease could reveal areas of dust emission, provided that areas of biomass burning are 12 excluded.

13 We used the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) annual Normalized 14 Difference Vegetation Index (NDVI) data set (Feldman and McClain, 2010) as a vegetation 15 proxy. The NDVI uses the normalized difference of the surface radiance in the red (absorbed 16 by chlorophyll) and in the infrared spectrum (scattered by the foliar structure) to indicate 17 vegetation abundance and structure (Brown et al., 2006). The choice of any specific NDVI 18 data set should have little impact on the source areas definition, as arid zones are the regions 19 with greater coherence between sensors (Brown et al., 2006) and we note that Behrenfeld et 20 al. (2001) reported high coherence of this data set with the legacy Advanced Very High 21 Resolution Radiometer (AVHRR) NDVI.

To promote spatial constraint, we used the spearman's ρ threshold of -0.9 -0.64 below which a pixel is identified as a source region. This is the minimum negative coefficient needed to attain statistical significance at the 0.05 level with all-eight years of data (2003-2010). The statistical significance is not an objective method to define source areas because a greater sample size would include areas with small correlation, but it is a convenient threshold for this first order approach.

To reduce the influence of biomass burning on the source areas definition, we further applied the constraint of a mean annual NDVI below 0.4. The and the results found here (see section 3.1), are markedly in agreement with other estimates of source areas in Patagonia. But biomass burning aerosols can be transported over large distances and could influence the AAI time series collected over the defined source regions. However, considering that Patagonia is 1 <u>a semi-desert environment is not likely that the AAI signal, especially over the source regions,</u>

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is significantly affected or determined by biomass burning events.

3 2.2 Phytoplankton response

4 The simplest and most commonly used parameter to remotely access phytoplankton responses 5 to environmental variability is the chlorophyll-a (Chl-a) concentration (Cropp et al., 2005), an universal pigment in the light harvesting process of photosynthesis (Ritchie, 2006). The fast 6 7 and pronounced Chl-a increase is a known effect following relief of iron limitation on 8 phytoplankton (de Baar et al., 2005), that is related to physiological acclimation and biomass 9 increase (Sunda and Huntsman, 1997; Westberry et al., 2013). Recently, Westberry et al. 10 (2013) used a series of remotely sensed proxies of phytoplankton response to revisit natural 11 and purposed iron fertilization studies in the last decade. In line with previous studies, their 12 results showed that although Chl-a concentration cannot be used reliable reliably to estimate 13 carbon stock changes, it is a sensitive parameter to iron addition. It could therefore be used as 14 a proxy to investigate phytoplankton response to dust derived iron inputs to the Southern 15 Ocean.

But for the dust fertilization hypothesis, leached iron additions should have an impact on the carbon stock, due to a delay in predators to cope with increased production (Irigoien, 2005) and the increase in abundance of larger, grazer-protected species. Therefore, we also evaluate changes in remotely sensed phytoplankton carbon (C_{phyto}) (Behrenfeld et al., 2005; Westberry et al., 2008, 2013) used as a biomass proxy for primary producers. C_{phyto} is derived from single band particulate backscattering (b_{bp}) at 443 nm, based on its modelled relationship with remotely sensed reflectance ($R_{rs}(443)$).

23 Diatoms are the primary group to respond to iron fertilization, but growth response is not 24 restricted to this group (Boyd et al., 2007). To evaluate which groups could be responding to 25 dust in this area, we also employed the relative annual frequency of broad groups from the 26 PHYSAT algorithm (Alvain et al., 2005, 2008). The PHYSAT algorithm uses 5 bands in the blue-green range of the visible electromagnetic spectrum to derive pixel dominance of 5 27 28 phytoplankton groups (Nanoeucariotes, Prochlorococcus, Synechococcus, Diatoms and Phaecystis-like) on a daily basis. Changes in the relative annual dominance frequency of these 29 30 groups could be related to dust variability.

1 Dust over the ocean or suspended in the seawater can interfere in carbon and pigment 2 readings by changing single band reflectance and the blue to green reflectance ratio used to calculate phytoplankton carbon stock and Chl-a from space-born sensors (O'Reilly et al., 3 2000; Moulin et al., 2001; Claustre et al., 2002). This could prevent the use of Chl-a and 4 5 C_{phyto} as proxies for dust mediated biological response. However, this effect should be minimal in waters where optical properties are dominated by biological constituents. The 6 7 mineral artefact effect is proportional to the relative concentrations of dust and 8 organisms/pigments (Claustre et al., 2002; Woźniak and Stramski, 2004), being more relevant 9 on oligotrophic areas and/or areas with high dust load in the atmosphere or in the seawater. Over the Atlantic sector of the Southern Ocean, dust load and flux are expected to be small 10 11 (Gaiero et al., 2003; Li et al., 2008) and the deep mixed layer in the region would decrease the 12 concentration of the dust deposited in the seawater (Claustre et al., 2002; De Boyer Montégut 13 et al., 2004). Phytoplankton biomass and pigment concentration however, are moderate to 14 high throughout the year (Allison et al., 2010). Combined, these properties minimize the noise 15 added by dust variation, suggesting a negligible effect of dust on biological proxy estimation in this region (e.g., Johnson et al., 2011). 16

17 Level 3 SeaWiFS annual Chl-*a* and GSM $b_{bp}(443)$ data (Feldman and McClain, 2010) from 18 2003 to 2010 were obtained, processed (C_{phyto}) and aggregated to 1°x1° spatial resolution. 19 Level 3 monthly relative frequencies from PHYSAT were aggregated to annual frequencies 20 and to 1°x1° spatial resolution. These data sets were then correlated <u>using Spearman's ρ with</u> 21 the AAI time series over the source areas in Patagonia. <u>Uncertainty estimates of the</u> 22 correlation analysis are presented in the supplementary material.

All analyses were carried out in R software (R Core Team, 2013) with aid of packages
"raster" (Hijmans and Etten, 2013), "rgdal" (Bivand et al., 2013a) and "sp" (Pebesma and
Bivand, 2005; Bivand et al., 2013b).

26 **3 Results and discussion**

27 **3.1** Source areas and interannual variation on dust emission

Four possible source areas were identified in Patagonia and Tierra del Fuego Island (Fig. 1). The major areas (i.e., Areas 1 and 2, Fig. 1) are located in northern and central Patagonia, on a sparsely vegetated region (Paruelo et al., 1998) punctuated by a disperse group of ephemeral rivers and lakes that provide wind erodible material (Prospero et al., 2002; Gaiero et al., 2003). Visual examination of the Moderate Resolution Imaging Spectroradiometer (MODIS)
 true colour composites (not shown) suggests that only a few point sources within these areas
 are major contributors to dust emission. The majority of the point sources are minor
 contributors, collectively compounding a diffuse brown haze over the southwest coast of the
 Atlantic Ocean, and only rarely presenting major emission events.

6 The major areas identified here are very similar to those published by Prospero et al. (2002), 7 which used AAI data in a different classification scheme, and are closely related to the areas 8 identified by the more recent remote sensing survey of Ginoux et al. (2012). Areas 1 and 2 9 also correspond to areas identified by the model of Johnson et al. (2010) as the major source 10 areas in Patagonia. Modelled source areas identified by Li et al. (2010) are somewhat similar, 11 but their southern area is centred around 50°S, over the San Julian Great Depression, which 12 showed only weak correlation in this study (Fig. 1).

13 The area in north-eastern Tierra del Fuego (Area 3, Fig. 1) has been previously described by 14 Arche and Vilas (1986, 2001) and recently proposed by Gassó et al. (2010) as the source for 15 mineral dust arriving at the Concordia Station (75.1°S, 123.35°E), Antarctica. Although small, 16 it is the southernmost recognized dust source in the Southern Hemisphere. This site is located 17 surrounding the San Sebastián bay, and is composed of seasonal dry lakes with strong wind 18 erosion patterns (Arche and Vilas, 1986, 2001). Dust plumes from this site can be easily 19 identified in MODIS true colour composites as elongated brown shades, due to the 20 combination of strong westerlies south of the Andes and aggregated deflation areas.

The area in western Tierra del Fuego (Area 4, Fig. 1), however, probably represents a miss identification by the procedure employed, as this site does not hold characteristics needed for dust emission. The islands surrounding the Beagle Channel represent a mixture of exposed rocks, forests and ice caps, without significant sedimentary sites. Therefore, this site is excluded from further processing.

As the simple arithmetic mean is sensitive to even a single extreme event, we computed the 10% trimmed mean AAI over the source regions. As expected due to area relations, AAI time series over these regions (Fig. 2) show dominance from areas 1 and 2. As dust emission could not be confirmed for area 4, time series used for further processing included only areas 1, 2 and 3. We note that this choice should exert minimal influence due to the close similarity between the time series.

3.2 Source activity and phytoplankton proxies correlation

2 The correlation of the mean AAI time series and Chl-a show a clear zonal pattern of positive 3 correlation over the Atlantic sector of the Southern Ocean downwind of Patagonia (Fig. 3A 4 and B). This zonal area of positive correlation is bounded by the limits of the Antarctic 5 Circumpolar Current Southern Boundary (ACC - SB) and the Subtropical Front (STF) at the 6 north, but generally restricted at the north of the Polar Front (PF; Fig. 3A), as defined by Orsi 7 et al. (1995). Although the zonal pattern is also visible in the C_{phyto} and AAI correlation (Fig. 8 3C and D), it is less clear and restricted to western areas closest to source, but with the same 9 zonal relations.

10 The correlation presented is an indirect analysis of dust interaction with the Southern Ocean 11 biological system, as the dust transport and deposition could not be evaluated. The patterns of 12 spatial transport depend heavily on the relative position of source areas and high/low pressure 13 zones (Johnson et al., 2011), while deposition also depends on the variable wet removal of dust particles from the atmosphere (Jickells et al., 2005). Nevertheless, spatial patterns of 14 15 annual dust deposition are highly coherent among models in this region, processed for 16 different years or periods (e.g., Mahowald et al., 2005). Together with the result showed here, 17 this suggests that on an annual scale, spatial variations in atmospheric dust transport and 18 deposition could be less important than dust source area activity. This could result because the 19 majority of the dust leaving the continent should deposit over the ocean and be redistributed by ocean currents, as it was estimated by Li et al. (2010) that only 13% of air masses leaving 20 Patagonia reach Antarctica within 10 days. 21

Notwithstanding the indirect analysis, the resulting zonal pattern is suggestive as it bears resemblance with the Southern Ocean meridional zonation in physical, chemical and biological features (Deacon, 1982; Pollard et al., 2002). The general pattern in the Chl-*a* correlation and its relation to the PF are also coherent with the results of Erickson et al. (2003), who used spatially resolved modelled dust deposition and monthly anomaly in Chl-*a* data between 2000-2001.

As areas of enhanced Chl-*a* are related to greater availability of iron on a first order basis (Sokolov and Rintoul, 2007), climatological distribution of Chl-*a* could be used as indirect assessment of nutritional status. Also, Chl-*a* absolute value is one important component of the artefact effect magnitude (Woźniak and Stramski, 2004). It is therefore valuable to analyse the relation between the climatological values of Chl-*a* (1997-2010) and the correlation index. This analysis shows lack of relation (Fig. 4), suggesting small to null system scale effect of nutritional (availability of trace metals) or artefact aspects (dust interference on the Chl-*a* signal) on the correlation. We also note that no clear longitudinal gradient in correlation strength along the zonal pattern is observed in Figs. 3A and B, which suggests also a lack of relation on dust flux and correlation indices, as observed by Erickson et al. (2003). As will be discussed later, this could result from a great sensitivity to even small additions of trace metals and the long range transport of dust particles suspended in the surface waters.

8 The similarity of the diatom annual relative dominance and dust source activity correlation 9 pattern (Fig. 5a) with those of Chl-*a* and C_{phyto} , and its association with the meridional zone 10 between the STF and the PF suggests a determinant role of large scale circulation and 11 biogeochemical features related to diatom ecology. This is expected, as diatoms are the 12 primary group limited by trace metals and Si availability in the Southern Ocean (Hutchins et 13 al., 2001; Leblanc et al., 2005).

Under prevailing limiting iron conditions of the Southern Ocean, the Si:NO₃ consumption 14 15 ratio can reach ~3:1, 2-3 times higher than in iron replete systems (e.g., Hutchins and 16 Bruland, 1998). The higher consumption ratio results in the preferential removal of Si relative 17 to other macronutrients from the surface waters (Hutchins and Bruland, 1998; Takeda, 1998). 18 With the advance of the growing season, shallow and more stratified waters north of the PF in 19 the Atlantic sector prevent sufficient resupply of Si from deeper waters (Sarmiento et al., 20 2004). Also, as water masses are advected northerly through the Ekman transport from the 21 upwelling site near continental Antarctica to the PF (Sigman et al., 1999), Si is removed by 22 the same process, resulting in advected waters with deficiency in Si relative to other 23 macronutrients (Hutchins and Bruland, 1998). Therefore, in contrast with waters south of the 24 PF, this region suffers a seasonal depletion of silicate in summer, reaching the limiting values for diatom growth of $< 5 \mu$ M (Coale et al., 2004; De La Rocha and Passow, 2004), an Si:N <25 0.5:1 (Garcia et al., 2010). The effect of such conditions is clear on late summer, when 26 27 communities in this region are typically dominated by non-diatom groups as both revealed by fraction of biomass basis (Laubscher et al., 1993) or pigment concentration (Alvain et al., 28 29 2008).

Upon iron addition, however, despite low Si availability, diatom assembly Chl-*a* and biomass
can increase several fold (Hutchins et al., 2001; Leblanc et al., 2005). The result is a reduced
Si:Diatom biomass and a community Si:NO₃ consumption that can drop below unity, to

average values of ~0.5:1, reducing the remaining Si to submicro-molar levels (Takeda, 1998; Coale et al., 2003; Leblanc et al., 2005). It is noteworthy that controls in waters very deficient in Si (< 0.6 μ M) have Si:NO₃ consumption ratio even smaller than Fe amended treatments (Hutchins et al., 2001; Leblanc et al., 2005). While Si is mainly consumed by diatoms, NO₃ can be consumed by other taxa, and the very low consumption ratio in the controls is due to a relatively low diatom growth compared with non-diatoms groups.

7 The mechanism behind the Fe effect in the Si:Diatom biomass and Si:NO₃ consumption ratio 8 is not yet clear and could involve synergic interactions of physiological (growth rate), 9 morphological (silification and/or volume) and taxonomical changes in the diatom assembly 10 (e.g., Marchetti and Cassar, 2009). Nevertheless, these effects are consistent and can be 11 observed even with small additions of 0.2-0.5 nM Fe to these waters (Coale et al., 2003; 12 Leblanc et al., 2005). Therefore, a possible dust fertilization effect could extend the growing season for diatoms, which would reduce the relative annual dominance of successional groups 13 like Synechoccocus (Alvain et al., 2008), as can be observed in Fig. 5b. 14

15 Dust dissolution could supply other trace metals to the surface layer, but their effects on 16 diatoms ecology are not yet clear. Under Si limitation, Zinc (Zn) additions can increase the 17 affinity of the Si uptake system, as measured by the lower half saturation constant for Si 18 uptake (De La Rocha et al., 2000), providing competitive advantage at least for some species 19 (Leblanc et al., 2005). But community experiments are variable in the effect of this element 20 on diatom growth and Si:NO₃ consumption ratios (Coale et al., 2003; Crawford et al., 2003; 21 Leblanc et al., 2005). Nevertheless, it is interesting that Zn concentrations are very low on the 22 area of high correlation (Wyatt et al., 2014) and that Croot et al. (2011) noted that Zn removal 23 occurred slightly after Si removal in meridional transects in the Atlantic sector of the 24 Southern Ocean.

But the plausibility of the dust fertilization hypothesis can be accessed for the iron effect alone. Assuming the average iron composition and solubility of Patagonian dust (Gaiero et al., 2003), it would be required a dust flux of 0.4 g m⁻² to add 0.5 nM Fe to a 50 m mixed layer depth, a value one order of magnitude lower than that estimated for singular dust events in Patagonia (0.2 to 2 mg m⁻²; Johnson et al., 2011). This value is also much smaller than would be needed for an artefact effect in the region. Considering the calculations of Woźniak and Stramski (2004) and an average mixed layer depth of 50 m, it would be required a dust flux of <u>5 g m⁻² for detectable artefact above the biological signal (0.1 g m⁻³ of dust for an average</u>
 chlorophyll concentration of 0.5 mg m⁻³).

Therefore, on an annual basis, increased addition of "background" dust or increased 3 4 frequency and intensity of dust events could provide measurable biological response within 5 the gradient of iron deficiency to iron replete systems on low Si waters at least for Chl-a. 6 Higher iron additions would be needed for biomass increase and the exponential decay on 7 atmospheric flux of mineral particles with distance from source could explain the response 8 restricted to western areas. Thus, the combined constraints of at least Fe and Si on diatom 9 growth possibly condition this region to be highly sensitive to dust derived iron additions, 10 explaining the spatial patterns. Direct effect of silicate addition from dissolution of dust in 11 seawater is unlikely (Boyd et al., 2010), as soluble Si represents up to 1% of aluminum-12 silicate mass (Tegen and Kohfeld, 2006), only two fold higher than iron in Patagonian dust (Gaiero et al., 2003), but with 10³ greater biological demand. 13

14 Examination of true colour composites reveals that thick Patagonian dust plumes disperse 15 being no longer visible after a few hundred kilometres from coast and therefore direct aeolian 16 input of biologically significant amounts of trace metals would be difficult on far off sites. 17 However, due to the integrity and dynamism of the fronts structure (Sokolov and Rintoul, 18 2007) and possible long residence time of dust in the mixed layer (Boyd et al., 2010), a 19 fraction of the dust deposited on the western region could be transported to distant sites, as 20 has already been suggested for Argentinean shelf sediments (de Baar et al., 1995). Also, 21 oceanic communities limited by micronutrients are efficient recyclers of iron. In such 22 communities, dust derived organically bounded iron can be transported hundreds of 23 kilometres from the addition location, as is observed downstream from islands (Sokolov and 24 Rintoul, 2007). This longitudinal transport by ocean currents would also help to explain the observed zonal pattern, not entirely coincident with modelled dust deposition patterns. 25

Finally, Fig. 3 also shows a strong positive relation at the blooming region along the Argentinean shelf-break. This region potentially receives micronutrients from upwelling, shelf water and direct dust deposition (Garcia et al., 2008). Shelf water micronutrients originate from a mixed contribution of which, apart from ground water discharge, aeolian dust can contribute to a minimum of ~40% (Gaiero et al., 2003). This indicates that interannual dust variation could influence an important fraction of the micronutrients delivered to this region. Although the correlation could be an artefact of wind influence on both upwelling and dust emission (Meskhidze et al., 2007) due to the close proximity of the sites, it need not be the case as dust variation can be related to wind-independent soil moisture and vegetation variation. As direct correlation on wind speed and Chl-a anomalies are positive for this region (Kahru et al., 2010), effects of these sources cannot be easily separated without models or field campaign, but we note that the two processes can occur simultaneously and need not be exclusive.

7 The results of Meskhidze et al. (2007) also cover the entire Atlantic sector of the Southern
8 Ocean, and show no evidence of direct effect of wind induced surface mixing on the
9 longitudinal band of correlation between dust and biological proxies. Nevertheless, it cannot

- 10 be excluded that the correlation observed is caused by external phenomenon imposing similar
- 11 variability to both dust and biological proxies, without a direct effect of dust fertilization. We
- 12 <u>are not aware, however, of such process.</u>

13 It is more difficult to understand the negative correlation surrounding the areas of positive 14 correlation. Air masses leaving the principal source regions in Patagonia potentially cover all Atlantic sector of the Southern Ocean (Li et al., 2010), from the Drake Passage to Southern 15 16 Africa, but are generally directed to southeast. Therefore negative (or positive) areas north of 17 ~40°S may have no relation with coupled dust and biological variability. South of 60°S, sea 18 ice dynamics and influence on biological communities would result in a lack of relation or at 19 least a small positive correlation due to dust accumulation on sea ice and subsequent release 20 in the same integration period (calendar year). However, areas south of ~60°S are also 21 generally negative, and the mechanism behind this behaviour is not yet understood.

22 4 Conclusions

The effect of Patagonian dust on the productivity of Atlantic sector of Southern Ocean has generally been regarded as negligible. Unfortunately, no research team has reported a dust storm from Patagonia over the ocean, so that the magnitude of dust flux in those conditions and its associated biological response are still unknown. Therefore, until observational or experimental data are available, we have to rely only on indirect studies based on modelling and remote sensing. The present study is an attempt to contribute with further indirect evidences.

30 Here we presented patterns of dust source activity correlation with three remotely sensed 31 proxies for biological response (chlorophyll-*a*, phytoplankton carbon concentration and 32 diatom relative dominance). The results showed a coherent spatial structure within the 1 meridional zonation of the Southern Ocean, extending from South America to Africa. The 2 correlation is interpreted in terms of dust supply of trace metals to surface waters, and 3 represents new evidence that trace metal dissolution from dust deposition has an observable 4 biological effect even at modern flux. The sensibility of the area north of the Polar Front is 5 attributed to diatom co-limitation by silicate and iron, although synergic effects of other 6 micronutrient supplied by dust could also contribute.

The possibility that Patagonian dust could help to modulate the productivity in the Atlantic
sector of the Southern Ocean even at the modern low flux is important in face of the current
climate change and land-use effects on dust source emission in Patagonia.

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- 28

Figure 1. Final contour of identified "source areas" overlaid on the AAI x NDVI correlation map. Visually confirmed source areas are: (1) northern Patagonia, (2) central Patagonia, and (3) northeastern Tierra del Fuego; area 4, in western Tierra del Fuego, was not confirmed and is probably a miss identification (see text).

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Figure 2. Mean <u>annual SCIAMACHY</u> AAI time series over source regions.

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8 Figure 3. Spearman correlation Correlation of mean AAI over source areas in Patagonia and 9 Chl-a (A, B) and C_{phyto} (C, D) over the Atlantic sector of the Southern Ocean. A and C show 10 the ocean fronts delimiting the larger spatial extent of the correlations, while B and C 11 superimpose ocean fronts delimiting areas with higher correlations. Grey corresponds to areas 12 with < 4 years of phytoplankton data. The fronts positions were obtained from Harris and 13 Orsi (2008).

14

Figure 4. Density plot of Spearman correlation indices dependency on Chl-*a* for the Southern
Ocean (> 40°S). Colour scale show number of points per bin. Thick line shows a robust
locally weighted regression (Cleveland, 1979).

18

Figure 5. Correlation of mean AAI over source areas in Patagonia and Diatom (A) and
Synechococcus (B) relative abundances over the Atlantic sector of the Southern Ocean. Grey
corresponds to areas with < 4 years of phytoplankton data. The scale is the same of Fig. 3.