Dear Editor Jean-Pierre Gattuso,

Please find attached the final version of the manuscript **bg-2019-352** entitled: "**Coccolithophore biodiversity controls carbonate export in the Southern Ocean**". A detailed explanation of all the changes included in the manuscript were posted on the Interactive discussion of our manuscript and are listed below.

<div align="center">**************************</div>

Response to reviewer 1

We sincerely thank **Dr Griet Neukermans** (reviewer 1) for her positive and constructive comments on our manuscript that have helped to improve the paper. We have carefully considered all her comments and addressed each of them as outlined below.

R1-Cx : Referee comment, **R1-Rx:** authors response.

R1-C1: *This paper is a very useful and original contribution to our understanding of how coccolithophore diversity shapes carbonate export in the Southern Ocean based on time series of sediment trap data. The paper is a pleasure to read: very well written, well structured, comprehensive, clear, and concise, with high-quality figures, and in-depth discussion. I highly recommend publication of this work in Biogeosciences. Congratulations to the authors for this very nice piece of work. I only have a few very minor comments that may improve the paper.*

R1-R1: We sincerely thank reviewer #1 for the careful reading of our manuscript and constructive criticisms and comments that helped to improve the manuscript. We have carefully considered all her comments and have addressed each of her concerns as outlined below.

R1-C2: *P3L87-89: replace "satellite reflectance observations" with "ocean color satellite reflectance observations" to precise that it is the fraction of incoming VISIBLE and NEARINFRARED solar radiation that is reflected from the ocean surface. Add reference (Balch et al., 2005) (Gordon et al., 2001) at the end of the sentence. These are the NASA standard algorithms for PIC retrieval.*

R1-R2: Corrected according to reviewer 1's suggestion.

R1-C3: *P4L111: reference for representativeness is missing*

R1-R3: Please note that a detailed explanation of the representativeness of the SOTS and SAM sites was explained later in the text (section 2.2). In the new version of the manuscript we refer to section 2.2 in the line indicated by the reviewer.

R1-C4: *P5L132: remove "that".*

R1-R4: Corrected according to reviewer 1's suggestion.

R1-C5: *P6 Figure 1: STZ not included in legend.*

R1-R5: Subtropical Zone **-** STZ has been included in the legend following reviewer 1's suggestion.

R1-C6: *P8L234: Can you briefly explain the method to calculate daily fluxes?*

R1-R6: The method employed to estimate coccolith and coccosphere fluxes has been included in the new version of the manuscript (lines 260-267 of the corrected version of the manuscript).

R1-C7:*P9L253-255. I strongly appreciate the authors obtained two independent estimates of coccolith fluxes based on the birefringence and morphometric methods, each with their own advantages and disadvantages.*

R1-R7: We appreciate reviewer 1's supportive comment. Since both techniques have associated errors, we decided to present both estimates (in the manuscript). Interestingly, in spite of some variability between techniques the general conclusions would remain similar to using any of the techniques individually.

R1-C8:*P10L294: Can you briefly explain why you think that the finding of <5% error on DSL estimates from polarization would apply to other species than the one tested?*

R1-R8: *E. huxleyi* overwhelmingly dominated the coccolithophore assemblages in all the samples analysed. Given the very low number of coccoliths of the rest of the coccolithophore species, it was almost impossible to find a representative number of individuals of for most of the "secondary" species in the same sample in order to statistically compare both microscopy techniques. Please note that even when a coccolith of a given species is found under the SEM, it can not always be measured because its position is not always adequate (e.g. they are often tilted or partially covered by other phytoplankton or detritus). Based on this, we decided to measure *C. leptoporus* because it was the second most abundant species, and therefore, statistical comparison between populations measured under the Light Microscope (LM) and SEM was possible. The subtle differences between coccolith distal length measurements are most likely due to the fact that the peripheral limit of the coccolith shield is not as sharp under the LM as is the case for SEM images. It follows that differences in coccolith measurements between SEM and LM techniques will be probably similar or smaller in the case of larger species. Since the majority of coccolith species identified in the current study display a similar (e.g. *Gephyrocapsa oceanica*, *Syracosphaera pulchra*, *Umbellosphaera tenuis* and *Umbilicosphaera sibogae*) or larger size (e.g. *Coccolithus pelagicus* and *Helicosphaera carteri*) than *C. leptoporus*, it can be assumed that the *<5% error on DSL estimates* for *C. leptoporus* is applicable to the rest of the species found in the current study. All the above is explained in section 2.6, lines 329-340 of the corrected version of the manuscript.

R1-C9: *Materials and Methods section: I think you should add a section on the ocean colour satellite data treatment. Which data did you use? Figure 2 suggests you used weekly data for PIC but monthly for Chla? Why not the same temporal resolution? Did you use multisensor merged products (such as GlobColour?)? Did you do any spatial averaging and how did you compute the weekly averages?*

R1-R9: Corrected according to reviewer 1's suggestion. A new subsection called "2.8 Remotely sensed chlorophyll-a and PIC concentrations" has been included in the new

version of the manuscript describing how we obtained and processed the Chl-*a* and PIC satellite data used in the manuscript. Weekly Chl-*a* data is now plotted in the graphs. Additionally, in order to support our statements in section 4.1 of the discussion, $CaCO_3$ fluxes registered by the traps have also been included in Figure 2.

R1-C10: *P12 Figure legend: specify "ocean color satellite-derived". Panel b, please add Chla data for October/November to see the potential rise in Austral spring. Can you present PIC and Chla data at the same temporal resolution? That would make sense.*
R1-R10: Corrected according to reviewer 1's suggestion. As mentioned in the previous comment weekly Chl-*a* data is now plotted in Figure 2. Moreover, data for the month of November is now included in figure 2.

R1-C11: *P12 Figure 2: panel c at 61S is missing.*

R1-R11: The figure caption erroneously mentioned the 61°S site (the figure caption corresponds to an earlier version of the manuscript where data from the 61°S was presented in the graph). In the new version of the manuscript this information is not required. Therefore, the reference to the 61°S site in the caption of Figure 2 has been deleted.

R1-C12:*P16L429: the secondary maximum of satellite PIC might be an artefact of satellite data treatment, but it's hard to say, since that critical information is missing from the manuscript Materials and Methods…*

R1-R12: As mentioned above (see **R1-R9**), a new subsection called "2.8 Remotely sensed chlorophyll-*a* and PIC concentrations" has been included in the manuscript. It is important to note that the PIC satellite signal for the grid area considered representative of the SAM station (coordinates 47-45° S and 171°E-179°W) was almost identical to that of a smaller area over the SAM site (47-45° S, 177.5-179.5°E). An alternative explanation of the secondary PIC maximum (i.e. possibility of storm-induced bubbles) has been included in the text. See section 4.1 of the new version of the manuscript.

R1-C13: *P14L377: Not clear what you mean with total CaCO3 export in Fig. 5. Is this the combined export of coccos and forams? If yes, how did you quantify foram export? I suggest you also explain total CaCO3 in the Figure legend.*
R1-R13: Both figure and figure caption have been modified in order to make clear that annual total $CaCO_3$ export (represented by yellow bars in Figure 5) refers to the total amount of $CaCO_3$ collected by the traps determined chemically (as explained in section 2.4) while the clear and dark blue bars represent the two different estimates of the contribution of $CaCO_3$ based on birefringence and morphometric techniques, respectively.

R1-C14: *P17L436: it may also be a foraminiferan signal, see for example (Rembauville et al., 2016).*

R1-R14: We appreciate reviewer 1's suggestion. Indeed, we did consider the possibility that heterotrophic calcifying plankton such as planktonic foraminifera or pteropods could account for the secondary maximum in February-early-March. However, total $CaCO_3$ fluxes recorded in the trap do not reflect an increase during this interval. Therefore, we

believe this possibility is unlikely. Please note that in the new version of the manuscript total CaCO$_3$ fluxes have been included in figure 2 (see also **R1-R9**).

R1-C15: *P18L497 etc.: The satellite PIC algorithm has indeed been calibrated in Northern hemisphere waters, where E. huxleyi greatly outnumbered other coccolithophore species, which is also the case in your study areas. In fact, the satellite signal (which is proportional to the particulate backscattering coefficient) is more sensitive to the concentration of E. huxelyi-sized particles, compared to larger, less abundant cocco species. Indeed, if larger, much heavier species are more prevalent in the Northern hemisphere waters, where the conversion factor for backscatter to PIC is calibrated, then this would lead to an overestimation of PIC in any waters where larger species are less prevalent. Put in other words, the conversion factor of backscatter to PIC is dependant on the size of the calcite particles. An alternative explanation for the overestimation of PIC is Southern Ocean waters is the contribution of bubbles to the backscattering coefficient.*

R1-R15: Our intention was to highlight that the different composition of coccolithophore assemblages between the Northern Hemisphere and Southern Ocean may contribute (only one factor among probably many) to the overestimation of PIC concentration in the Southern Ocean. In the new version of the manuscript this has been clarified and the possible influence of microbubbles to the backscattering coefficient has also been included.

R1-C16: *P23L654: poleward expansion of E. huxleyi to the Arctic has also been demonstrated by (Neukermans et al., 2018)*

R1-R16: We appreciate the new reference provided by reviewer 1. This study is now mentioned in the new version of the manuscript.

R1-C17: *P24L664 etc.: see also recent review in (Krumhardt et al., 2017)*

R1-R17: The reference mentioned by reviewer 1 has been included in the discussion (section 4.5).

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Response to reviewer 2

We sincerely thank **Dr Alex Poulton** (reviewer 2) for the valuable comments and suggestions that have helped to improve the original version of the manuscript. We have carefully considered all his comments and have addressed each of his concerns as outlined below.

R2-Cx : Referee comment, **R2-Rx:** authors response.

R2-C1: *The manuscript by Rigual Hernandez et al. represents a comprehensive study of species-specific fluxes of coccolithophore-derived CaCO3 fluxes to the deep-sea in the*

Southern Ocean. The manuscript is well written and easy to follow, and provides several new insights into the important role of numerically rare coccolithophore species with high relative coccolith and cellular CaCO3 content. Such understanding has been well recorded in terms of production and export in northern polar and sub-polar waters, but the manuscript by these authors reveals the importance of this processes in the Australian-New Zealand sector of the Southern Ocean. There are no significant issues with the methods or conclusions, and only a few points that need clarity or further referencing.

R2-R1: We sincerely appreciate reviewer 2 for taking the time to carefully read the manuscript and providing valuable comments and references that helped to improve the manuscript.

R2-C2: *Ln 39: 'E. huxleyi dominates remote sensing images as a result of higher cell abundance and detachment of its small coccoliths.' This is an oversimplification and ignores the vital role of the characteristic light-scattering properties and size of E. huxleyi coccoliths, in addition to its tendency to shed coccoliths and characteristic bloom formation.*

R2-R2: The sentence referred by reviewer 2 has been replaced by the following: "This observation contrasts with the generally accepted notion that high PIC accumulations during the austral summer in the subantarctic Southern Ocean are mainly caused by *E. huxleyi* blooms.".

R2-C3: *Ln 56-57: 'decline in saturation state of carbonate minerals in seawater makes the biological precipitation of carbonate difficult and increases dissolution rates of their shells or skeletons'. Current theoretical consensus of the response of coccolithophores to carbonate chemistry (e.g. Bach et al., 2015) specifically relates their internal calcification to substrate availability (HCO3-) and inhibition by proton (H+) concentrations; i.e. different carbonate chemistry parameters than inferred in the text (i.e. CaCO3 saturation state).*
Bach et al. (2015). A unifying concept of coccolithophore sensitivity to changing carbonate chemistry embedded in an ecological framework. Progress in Oceanography, 135, 125-138.

R2-R3: The sentence highlighted by Reviewer 2 has been modified taking into consideration his suggestion and the reference of Bach et al. (2015) is now mentioned in the text (see lines first part of the introduction of the corrected version of the manuscript). Please note that in this part of the introduction we are talking in general about marine calcifying organisms, i.e. not specifically about coccolithophores.

R2-C4: *Ln 92-95: As well as recent work by Trull et al. (2018) showing that satellite oceancolour based PIC estimates can be unreliable in Antarctic waters, should also cite Holligan et al. (2010) which came to the same conclusion earlier.*

R2-R4: Corrected according to reviewer 2's suggestion. Holligan et al. (2010) paper is now mentioned together with Trull et al. (2018) in the new version of the manuscript.

R2-C5: *Ln 131-132: 'which that', delete one or the other, both not necessary.*

R2-R5: Corrected according to reviewer 1 and 2's suggestion.

R2-C6: *Ln 294-295: 'For the ks value of each taxa, data from the literature were (Table 1)' – sentence not finished.*

R2-R5: The sentence referred to by reviewer 2 has been modified. Now it reads: "For the k_s value of each taxa, data from the literature was employed (Table 1)."

R2-C7: *Ln 329: Missing word – 'later' at end of sentence 'i.e. approximately eight months <later> (Fig. 2).'*

R2-R5: We intended to say that the period of elevated coccolith flux lasted about 8 months. However, this information is not of critical importance and therefore we have deleted the end of the sentence.

R2-C8: *Fig. 2. Would it not be better to make the y-axis on these plots the same scale?*

R2-R5: Corrected according to reviewer 2's suggestion. The y-axes have now the same scale in each station. Please note that in figure SAM site two axes (coccospheres and PIC) required different scale due to the different magnitude of these parameters compared to those of the SOTS site.

R2-C9: *Ln 417-419: This is an interesting point, as it is similar to loss terms found specifically for coccolithophores from microzooplankton grazing in the temperate N Atlantic setting (60-80%; Mayers et al., 2019).*
Mayers et al. (2019). Growth and mortality of coccolithophores during spring in a temperate Shelf Sea (Celtic Sea, April 2015). Progress in Oceanography 177, 1010928.

R2-R9: We appreciate reviewer 2's suggestion. This is a good point that has been included in the new version of the manuscript (see section 4.1 of the new version of the manuscript).

R2-C10: *Ln 490-492: Again, although Trull et al. (2018) recently identified over-estimate of coccolithophore PIC in the Southern Ocean by the NASA satellite ocean-colour-based PIC algorithm, this was examined earlier by Holligan et al. (2010). In the case of Holligan et al. (2010), the difference was attributed to the lower coccolith and cell CaCO3 content of E huxleyi found in the S Atlantic (Scotia Sea). This is in general agreement with the reasoning suggested here (i.e. issues over the coccolith specific-area:mass ratios for the dominant reflective particles), though differs over whether this is considered a problem with E huxleyi or C pelagicus (or other species with high coccolith CaCO3 content).*

R2-R10: We agree with reviewer 2. The text has been modified including Holligan et al. (2010) reference in the manuscript. Now it reads: "Since satellite reflectance observations are mainly calibrated against Northern Hemisphere PIC results (Balch et al., 2011; Balch et al., 2016), the lower the calcite content of dominant E. huxleyi morphotypes (B/C) in the Southern Ocean compared to their northern hemispheric counterparts has been suggested as a possible factor causing the over-estimation of PIC concentrations in the Southern Ocean. Following this reasoning, we speculate that differences in other components of the coccolithophore assemblages, and particularly, differences in C.

pelagicus numbers, could contribute to the over-estimation of PIC concentrations by the satellite PIC algorithm in the Southern Ocean. Indeed,…"

R2-C11: Ln 570: Should the units not be 0.4 Tmol C yr-1?

R2-R11: Reviewer 2 is correct, this error has been corrected in the new version of the manuscript.

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Additional corrections

Additionally, a few extra minor changes have been included in the text and are listed below:

- Some small changes have been included in results section 3.2. The annual relative abundance of coccolithophore species of the SOTS site has been updated. The changes only affect secondary species and are almost negligible (~0.1%). Moreover, some coccolithophore species with very low contribution but present in the samples of the SAM station were not listed in the first version of the manuscript. However, we believe all species should be mentioned in the text and therefore have been included all of them (see las sentence of section 3.2). Please note that none of these changes have an influence on the rest of the results or discussion.

- Sentence on Line 754. The reference of Rintoul et al. (2018) has been deleted and the sentence slightly modified. This publication does not fully support our statement and therefore we believe it is more appropriate to delete it from the text.

- Both reviewers are now thanked in the in the acknowledgment section.

- All data has been uploaded in the Australian Antarctic Data Centre in the following link: https://data.aad.gov.au/metadata/records/Coccolithophore_Fluxes_SAZ_2009-2012

1 **Coccolithophore biodiversity controls carbonate export in the Southern Ocean**

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21 <u>**Abstract**</u>

22   Southern Ocean waters are projected to undergo profound changes in their

23 physical and chemical properties in the coming decades. Coccolithophore blooms in the

24 Southern Ocean are thought to account for a major fraction of the global marine calcium

25 carbonate ($CaCO_3$) production and export to the deep sea. Therefore, changes in the

26 composition and abundance of Southern Ocean coccolithophore populations are likely to

27 alter the marine carbon cycle, with feedbacks to the rate of global climate change.

28 However, the contribution of coccolithophores to $CaCO_3$ export in the Southern Ocean is

29 uncertain, particularly in the circumpolar Subantarctic Zone that represents about half of

30 the areal extent of the Southern Ocean and where coccolithophores are most abundant.

31 Here, we present measurements of annual $CaCO_3$ flux and quantitatively partition them

32 amongst coccolithophore species and heterotrophic calcifiers at two sites representative

of a large portion of the Subantarctic Zone. We find that coccolithophores account for a major fraction of the annual $CaCO_3$ export with highest contributions in waters with low algal biomass accumulations. Notably, our analysis reveals that although *Emiliania huxleyi* is an important vector for $CaCO_3$ export to the deep sea, less abundant but larger species account for most of the annual coccolithophore $CaCO_3$ flux. This observation contrasts with the generally accepted notion that high PIC accumulations during the austral summer in the subantarctic Southern Ocean are mainly caused by *E. huxleyi* blooms. It appears likely that the climate-induced migration of oceanic fronts will initially result in the poleward expansion of large coccolithophore species increasing $CaCO_3$ production. However, subantarctic coccolithophore populations will eventually diminish as acidification overwhelms those changes. Overall, our analysis emphasizes the need for species-centred studies to improve our ability to project future changes in phytoplankton communities and their influence on marine biogeochemical cycles.

1. Introduction

The emissions of carbon dioxide (CO_2) into the atmosphere by anthropogenic industrial activities over the past 200 years are inducing a wide range of alterations in the marine environment (Pachauri et al., 2014). These include ocean warming, shallowing of mixed layer depths, changes in nutrient supply to the photic zone, and decreasing carbonate-ion concentrations and pH of the surface ocean, a process known as ocean acidification (Rost and Riebesell, 2004; Stocker et al., 2014). Substantial evidence from CO_2 manipulation experiments indicates that many species of corals, pteropods, planktonic foraminifera and coccolithophores will reduce their calcification rates under future ocean acidification scenarios (Bijma et al., 2002; Langdon and Atkinson, 2005 among others; Orr et al., 2005; Bach et al., 2015; Meyer and Riebesell, 2015). Owing to their moderate alkalinity and cold temperatures, Southern Ocean waters are projected to become undersaturated with respect to aragonite no later than 2040 and to calcite by the end of the century (Cao and Caldeira, 2008; McNeil and Matear, 2008; Shadwick et al., 2013). This decline in the saturation state of carbonate, together with other changes in carbonate chemistry speciation, will enhance dissolution of both aragonite and calcite shells and will make the biological precipitation of carbonate difficult in some marine calcifying organisms (Fabry et al., 2008; Gattuso and Hansson, 2011). Since such thresholds will be reached sooner in polar regions, Southern Ocean ecosystems have been

81 proposed as bellwethers for prospective impacts of ocean acidification on marine
82 organisms at mid and low latitudes (Fabry et al., 2009).

83   Coccolithophores are a major component of phytoplankton communities in the
84 Southern Ocean, particularly in its northern-most province, the Subantarctic Zone, where
85 they often exhibit maximum abundances and diversity (e.g. Gravalosa et al., 2008;
86 Saavedra-Pellitero et al., 2014; Malinverno et al., 2015; Charalampopoulou et al., 2016).
87 Coccolithophores play an important and complex role in the Southern Ocean carbon cycle
88 (Salter et al., 2014). On the one hand, the production of calcite platelets (termed
89 coccoliths) decreases the alkalinity of surface waters thereby reducing the atmospheric
90 uptake of CO_2 from the atmosphere into the surface ocean. On the other hand, the
91 production of organic matter through photosynthesis, and its subsequent transport to
92 depth in settling particles, enhances carbon sequestration via the biological carbon pump
93 (Volk and Hoffert, 1985). Additionally, due to their high density and slow dissolution,
94 coccoliths act as an effective ballast for organic matter, increasing organic carbon
95 sequestration depths (Buitenhuis et al., 2001; Boyd and Trull, 2007; Ziveri et al., 2007).
96 Therefore, changes in the abundance, composition and distribution of coccolithophores
97 could have an extensive impact on ocean nutrient stoichiometry, carbon sequestration,
98 and nutrition for higher trophic levels in the Southern Ocean (Deppeler and Davidson,
99 2017).

100   The remoteness and vastness of the Southern Ocean, together with the inherent
101 temporal and spatial variability of pelagic ecosystems, hampers accurate characterization
102 and quantification of Southern Ocean phytoplankton communities. Advances in satellite
103 technology and modelling algorithms have allowed a circumpolar and year-round
104 coverage of the seasonal evolution of major phytoplankton functional groups within the
105 Southern Ocean (e.g. Alvain et al., 2013; Hopkins et al., 2015; Rousseaux and Gregg,
106 2015). In particular, ocean-colour satellite reflectance observations have been used to
107 quantitatively estimate coccolithophore Particulate Inorganic Carbon (PIC)
108 concentrations throughout the Southern Ocean (Gordon et al., 2001; Balch et al., 2005b).
109 These satellite estimates suggest apparent high PIC values during summer near the major
110 Southern Ocean fronts attributed to coccolithophores (Balch et al., 2011; Balch et al.,
111 2016). This band of elevated reflectance and PIC that encircles the entire Southern Ocean
112 was termed the "Great Calcite Belt" by these authors. However, comparison of satellite
113 remote-sensing data with ship-based observations (Holligan et al., 2010; Trull et al.,

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2018) indicate, that satellite ocean-colour-based PIC estimates could be unreliable, particularly in Antarctic waters where they erroneously suggests high PIC abundances. Shipboard observations, on the other hand, provide a detailed picture of phytoplankton community composition and structure, but are dispersed, both temporally and geographically, and provide rather heterogenous data in terms of taxonomic groups investigated, and the sampling scales and methodologies used (e.g. Kopczynska et al., 2001; de Salas et al., 2011; Poulton et al., 2013; Patil et al., 2017, among others). *In situ* year-round monitoring of key strategic regions is critically needed to establish baselines of phytoplankton community composition and abundance and to validate and improve ocean biogeochemical models (Rintoul et al., 2012). This information is also essential if we are to detect possible climate-driven changes in the structure of phytoplankton communities that could influence the efficiency of the biological carbon pump, with consequent feedbacks to the rate of deep-water carbon sequestration and global climate change (Le Quéré et al., 2007; Deppeler and Davidson, 2017).

Here, we document coccolithophore and carbonate particle fluxes collected over a year by four sediment trap records deployed at two strategic locations of the Australia and New Zealand sectors of the Southern Ocean considered representative of a large portion of the SAZ (See section 2.2 for further details). Our measurements provide coccolith mass estimates of the main coccolithophore species and quantitatively partition annual carbonate fluxes amongst coccolithophore species and heterotrophic calcifiers. We find that coccolithophores are a major vector for $CaCO_3$ export out of the mixed layer and that the largest contribution to $CaCO_3$ export is not from the most abundant species *Emiliania huxleyi* but rather from larger coccolithophores species with substantially different physiological traits (e.g. *Calcidiscus leptoporus*). Our results emphasize the urgent need for diagnostic fitness response experiments on other coccolithophore species aside from *E. huxleyi* (e.g. Feng et al., 2017) in order to be able to be able to predict the impacts of anthropogenically induced changes in Southern Ocean ecosystems and biological carbon uptake mechanisms.

2. Material and methods

2.1 Oceanographic setting

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150   The SAZ alone accounts for more than half of the Southern Ocean area (Orsi et

151   al., 1995) and represents a transitional boundary between the warm, oligotrophic waters

152   of the subtropical gyres to the north and the cold, silicate-rich waters south of the Polar

153   Front (PF). The SAZ is arguably the largest high nutrient, low chlorophyll (HNLC)

154   province in the world's ocean and is central to the linkages between the ocean–

155   atmosphere CO_2 exchange and climate. The deep winter convection in the SAZ, which

156   exceeds 400 m, results in the formation of a high-oxygen water masses known as

157   Subantarctic Mode and Antarctic Intermediate Waters that connect the upper and lower

158   limbs of the global overturning circulation (Sloyan and Rintoul, 2001a, b). The formation

159   of these water masses are responsible for the sequestration of a large fraction of

160   anthropogenic CO_2 (Sabine et al., 2004), with an estimated 1 Gt C yr^{-1} transported to

161   intermediate depths annually (Metzl et al., 1999). Macronutrient concentrations display

162   pronounced seasonal changes in the SAZ with fully replete levels during winter to

163   substantial depletion during summer, particularly for silicate (Dugdale et al., 1995;

164   Rintoul and Trull, 2001; Bowie et al., 2011). Phytoplankton community in the

165   subantarctic zone is dominated by pico- and nanoplankton including cyanobacteria,

166   coccolithophores and autotrophic flagellates with lower abundances of diatoms than polar

167   waters south the Polar Front (Chang and Gall, 1998; Kopczynska et al., 2001; de Salas et

168   al., 2011; Rigual-Hernández et al., 2015b; Eriksen et al., 2018).

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2.2 Field experiments

171   Here we report on the coccolithophore and biogeochemical fluxes collected over

172   a year at the Australian Southern Ocean Time Series (SOTS) observatory (Trull et al.,

173   2010) and the New Zealand Subantarctic Mooring (SAM) site (Nodder et al., 2016) (Fig.

174   1). The SOTS observatory is located in the abyssal plane of the central SAZ

175   approximately 530 km southwest of Tasmania (46° 56' S, 142° 15' E) within an anti-

176   cyclonic gyre in a region characterized by weak circulation (Trull et al., 2001; Herraiz-

177   Borreguero and Rintoul, 2011). SOTS was equipped with three vertically moored, conical

178   time-series sediment traps (McLane Parflux Mk 7G-21) placed at ~1000, 2000 and 3800

179   m depth between August 2011 until July 2012. The physical, chemical and biological

180   parameters of SOTS site are regarded as representative for large portion of the Indian and

181   Australian sectors of the SAZ (~90°E and 140°E; Trull et al., 2001). The SAM site is

182   located in the Bounty Trough in in the subantarctic waters south east of New Zealand

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184 (46°40'S, 178' 30°E) and was equipped with conical, time-incremental sediment trap
185 (McLane PARFLUX Mk7G-21) placed at 1500 m depth, with samples used in the present
186 study collected between November 2009 until November 2010. The SAM site is
187 considered to be representative of a wide area of the northern sector of the SAZ off eastern
188 New Zealand, approximately 171°E to 179°W and 45 to 47°S (Law et al., 2014; Fig. 1).
189 Full details of the field experiments from these two localities in the Australian and New
190 Zealand sectors of the SAZ can be found in Trull et al. (2001) and Nodder et al. (2016),
191 respectively.

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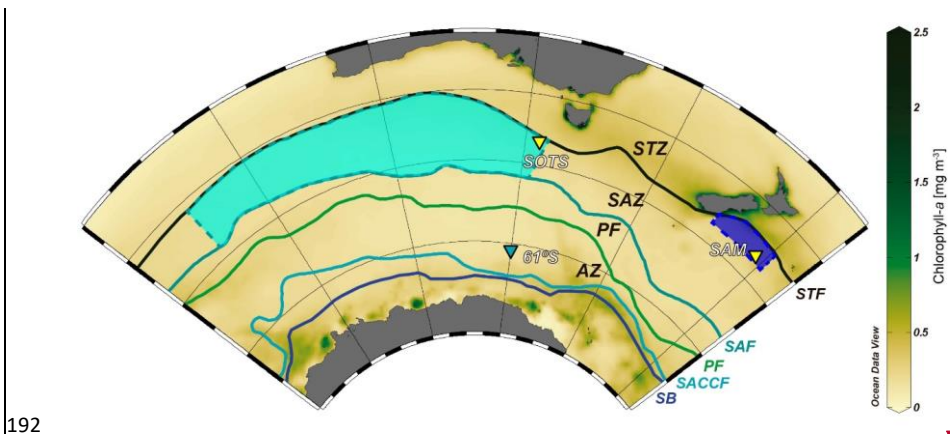
Figure 1: Chlorophyll-*a* composite map of the Australian-New Zealand sector of the
Southern Ocean (July 2002 to July 2012) from the MODIS Aqua Sensor showing the
location of the sediment trap moorings sites: SOTS, 61°S and SAM. The regions for
which the SOTS and SAM sites are representative are marked with light and dark blue
areas, respectively. Abbreviations: Subtropical Zone – STZ, Subtropical front - STF,
Subantarctic Zone – SAZ, Subantarctic Front - SAF, Polar Frontal Zone - PFZ, Polar
Front - PF, Antarctic Zone – AZ, Southern Antarctic Circumpolar Current Front –
SACCF, southern boundary of the ACC – SB. Oceanic fronts after Orsi et al. (1995).

2.3 Sample processing

In short, the recovered trap bottles were refrigerated upon recovery and then
allowed to settle. The sample slurry was then wet-sieved through a 1 mm screen in the
case of SOTS (no attempt to extract zooplankton "swimmers" was made for the <1 mm
fraction analysed here) and through a 200 µm sieve to remove "swimmers" for the SAM
site. The remaining fraction was then split using a McLane wet sample divider; the SOTS

6
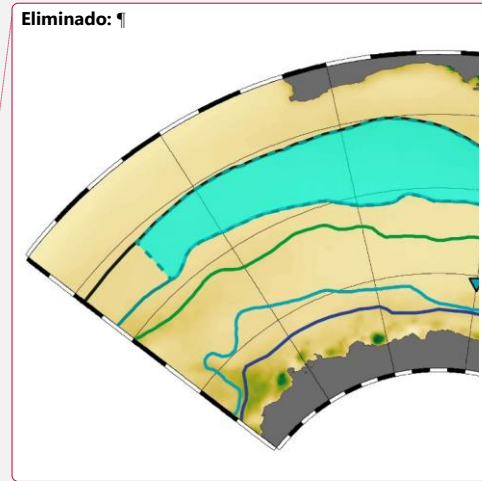
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samples were subdivided into one tenth aliquots while one fifth splits were made for the SAM samples. For the SOTS samples, a total of 55 samples were processed for calcareous nannoplankton analysis. The one-tenth splits dedicated to phytoplankton analysis were further subdivided into four aliquots with the McLane splitter. One aliquot was used for calcareous nannoplankton analysis and the remaining three were kept refrigerated for biomarker and non-calcareous microplankton analyses. In the case of the SAM samples, the one-fifth aliquots were further subdivided into five subsplits, and one of those was used for calcareous nannoplankton analysis. Two different types of glass slides per sample were prepared. The first preparation was used for the estimation of coccosphere and calcareous dinocyst (calcispheres of thoracosphaerids) fluxes and for coccolith imaging. A volume ranging between 1000 and 5000 µl of the raw sample was mounted on a glass slide using Canada balsam following Flores and Sierro (1997). This technique produces random settling of the coccoliths for microscopic identification and enumeration. The second type of glass slide was prepared following a modified protocol for non-destructive disintegration of aggregates modified from Bairbakhish et al. (1999). The objective of this chemical treatment is to reduce biases in the coccolith flux estimations associated with the presence of different types of aggregates and coccospheres (Bairbakhish et al., 1999). In brief, 2000 µl were extracted from the aliquot for calcareous nannoplankton analysis and then treated with a solution comprising 900 µl sodium carbonate and sodium hydrogen carbonate, 100 µl ammonia (25%) and 2000 µl hydrogen peroxide (25%). The sample was agitated for 10 seconds every 10 minutes and this process was repeated over an hour. Then, the reaction was stopped with catalase enzyme and samples were allowed to settle for at least 48 hours before preparation on microscope slides. pH controls indicate that the solution kept pH levels near 9, therefore precluding coccolith dissolution. Finally, trap samples were mounted on microscope slides following the same decantation method as used for the first type of glass slides (i.e. Flores and Sierro, 1997).

2.4 Determination of $CaCO_3$ fluxes

A detailed description of the geochemical analytical procedures for the SOTS samples is provided in Trull et al. (2001) and Rigual-Hernández et al. (2015a) while more detailed procedures of the SAM trap can be found in Nodder et al. (2016). In short, for the SOTS site three of the one tenth splits were filtered onto 0.45 pore size filters. Then the material was removed from the filter as a wet cake of material, dried at 60°C, and

ground in an agate mortar. This material was used to determine the total mass and composition of the major components of the flux. Particulate inorganic Carbon (PIC) content was measured by closed system acidification with phosphoric acid and coulometry. For the SAM site, one-fifth split was analysed for elemental calcium (Ca) concentration using ICP-MS techniques. The samples were oven-dried, digested in nitric/hydrochloric acid and then analysed according to the methods under US EPA 200.2. Ca was used to estimate $CaCO_3$ content in the samples assuming a 1:1 molar ratio in $CaCO_3$.

2.5 Quantification and characterization of coccolithophore sinking assemblages

Qualitative and quantitative analyses of coccospheres and coccoliths were performed using a Nikon Eclipse 80i polarised light microscope at 1000 x magnification. The taxonomic concepts of Young et al. (2003) and the Nannotax website (Young et al., 2019) were used. A target of 100 coccospheres and 300 coccoliths was established; however, owing to the pronounced seasonality in coccolithophore export, there were some periods with very low abundance of coccospheres in the samples and therefore the target of 100 coccospheres was not always met. Coccosphere and coccolith species counts were then transformed into relative abundances and daily fluxes using the following formula:

$$F = \frac{N \times \frac{A}{n \times a} \times V \times S}{d \times T}$$

where F = coccolith flux, N = number of coccoliths, A = area of the Petri dish, n = number of fields of view, a = area of a field of view, V = dilution volume, S = sample split, d = number of days of collection and T = sediment trap aperture area.

2.6 Determination of coccolith mass and size

Birefringence and morphometric methods are the two most commonly used approaches for estimating the calcite content of isolated coccoliths. The circularly-polarized light-microscopy-based technique (Fuertes et al., 2014) is based on the systematic relationship between the thickness of a given calcite particle (in the thickness range of 0 - 1.55 mm) and the first-order polarization colours that it displays under

277 polarized light (Beaufort, 2005; Beaufort et al., 2014; Bolton et al., 2016). The advantages
278 of this approach are that: (i) it directly measures complete coccoliths with no assumptions
279 regarding their shape or thickness and (ii) it allows for quantification of calcite losses
280 associated with missing parts or etching of the coccoliths. Disadvantages of this technique
281 are the errors associated with the coccolith-calcite calibration and their consequent effect
282 on the coccolith mass estimates (Fuertes et al., 2014; González Lemos et al., 2018). The
283 morphometric approach, on the other hand, allows better taxonomic identification of the
284 coccoliths and has smaller errors in the length measurements (~0.1 to 0.2 μm; Poulton et
285 al. 2011). However, this method does not allow direct measurement of coccolith thickness
286 and assumes identical shape and width proportions for all specimens of the same species,
287 among other uncertainties (see Young and Ziveri, 2000 for a review). Since the two
288 methods have different associated errors (Poulton et al., 2011), we applied both
289 approaches to our coccolith flux data in order to obtain two independent estimates of the
290 fractional contribution of coccolithophores species to total carbonate export in the SAZ.

291   For the birefringence-based approach, a minimum of 50 coccoliths of each of the
292 main coccolithophore species were imaged using a Nikon Eclipse LV100 POL light
293 microscope equipped with circular polarisation and a digital camera (Nikon DS-Fi1 8-bit
294 colour). The only exception was *E. huxleyi* for which coccolith mass values had already
295 been estimated in all the same samples at high resolution by Rigual-Hernández et al.
296 (under review). For the minor components of the flux assemblage, a lower number of
297 coccoliths were measured (Table 1). A photograph of the same apical rhabdolith of the
298 genus *Acanthoica* was taken and used for calibration at the beginning of each imagining
299 session during which microscopy light and camera settings were kept constant. A
300 different number of fields of view of multiple samples representative of different seasons
301 were photographed until the target number of coccoliths for each species was reached.
302 Photographs were then analysed by the image processing software C-Calcita. The output
303 files for single coccoliths were visually selected and classified into the lowest possible
304 taxonomic level. Length and weight measurements were automatically determined by C-
305 Calcita software. Morphometric measurements of all the species are summarized in Table
306 1. For further methodological details see Fuertes et al. (2014) and Bolton et al. (2016).

307   The second approach consisted of performing morphometric measurements on the
308 coccoliths followed by the estimation of their coccolith mass assuming a systematic
309 relation between length and thickness (Young and Ziveri, 2000). Young and Ziveri (2000)

proposed that the calcite content of a given coccolith could be estimated using the following formula:

$$\text{Coccolith calcite (pg)} = 2.7 \times k_s \times l^3$$

where 2.7 is the density of calcite ($CaCO_3$; pg μm^3), "k_s" is a shape constant that varies between species and morphotypes and whose value is based on the reconstruction of coccolith cross profiles and "l" is the distal shield length (DSL). In order to undertake coccolith measurements on the same coccoliths used for the birefringence-based approach, we employed the distal shield length values measured by C-Calcita using circularly polarized light instead of morphometric measurements on Scanning Electron Micrographs (SEM) as made in Young and Ziveri (2000).

Since coccolith distal shield length (DSL) has been reported to be systematically underestimated using cross-polarized light microscopy (e.g. D'Amario et al., 2018), we evaluated the possible errors in the DSL measurements made by C-Calcita. For this assessment, we measured 40 detached coccoliths of *C. leptoporus* under the SEM from samples of the SOTS sediment traps using the image processing software Image-J. Average DSL measurements under the SEM were then compared with those made by C-Calcita on 40 randomly selected *C. leptoporus* coccoliths. The average coccolith length obtained with the SEM analysis (6.37 ± 1.02, $n = 40$) was ~ 4% shorter than that estimated with C-Calcita (6.62 ± 1.47, $n = 40$). Therefore, we assumed the error for the DSL measurements with circularly polarized light is < 5%. Given the low numbers of the rest of species in the samples we considered that this error is applicable for the rest of the taxa measured in the current study. The subtle differences in coccolith distal length measurements between techniques are most likely due to the fact that the peripheral limit of the coccolith shield under the circulary-polarized light microscope (LM) is not as sharp as is the case for SEM images. It follows that differences in DSL measurements between SEM and LM techniques will be likely similar or smaller in the case of larger species. Since the majority of coccolith species identified in the current study display a similar (e.g. *Gephyrocapsa oceanica*, *Syracosphaera pulchra*, *Umbellosphaera tenuis* and *Umbilicosphaera sibogae*) or larger size (e.g. *Coccolithus pelagicus* and *Helicosphaera carteri*) than *C. leptoporus*, it could be assumed that the <5% error on DSL estimates for *C. leptoporus* is applicable for the rest of the species found in the current study. For the k_s value of each taxa, data from the literature was employed (Table 1). *E. huxleyi* assemblages in the SAZ are composed of a mixture of five different morphotypes: A, A

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overcalcified, B, B/C and C, each of which is characterized by different shape factors (k_s). Since k_s is not available for all the morphotypes found in the SAZ and it is not possible to differentiate between morphotypes in our light microscopy images, we used the mean shape factor constant for *E. huxleyi* (i.e. $k_s = 0.0275$) to provide a range of coccolith mass estimates for this species (Table 1 and Fig. 4).

2.7 Calculation of annual estimates

Since the trap collection periods encompassed a period shorter than a calendar year, annual estimates of coccolith and $CaCO_3$ fluxes and species relative abundances had to be estimated. For the SOTS site, a total of 336 days were sampled for the 1000 and 2000 m traps and 338 days for the 3800 m. Since the unobserved interval occurred in winter, the missing sampling period was filled using an average flux value of the winter cups (first and last trap bottles). In the case of the SAM trap, the number of samples available for $CaCO_3$ and calcareous nannoplankton analyses was different, covering a period of 313 and 191 days respectively. Since gaps were quasi-equally distributed along the time series, annual fluxes were estimated by filling the gaps in the record with average fluxes calculated from the available data. The estimated range of the annual contribution of coccolithophores to total $CaCO_3$ export at the SOTS and SAM traps was calculated by multiplying the coccolith flux of each species in each sampling interval by its average coccolith weight values obtained with the birefringence and morphometric techniques.

2.8 Remotely sensed chlorophyll-*a* and PIC concentrations

Weekly Chlorophyll-a and PIC concentrations for the sampling intervals at the SOTS and SAM sites were derived from Giovanni online data system, developed and maintained by the NASA Goddard Earth Sciences Data Active Archive Center (Acker and Leptoukh, 2007). Each value is a weekly value is produced by computing spatial averages within the area 48.5-45.5°S and 130-150°E for the SOTS site and 47-45°S and 171°E-179°W for the SAM site (Fig. 5).

3. RESULTS

3.1 Magnitude and seasonality of coccolithophore and $CaCO_3$ fluxes

Annualized coccolith fluxes were similar at the SOTS three trap depths, with 8.6, 7.3 and 8.6 x 10^{11} liths m^{-2} yr^{-1} at 1000, 2000 and 3800 m respectively, and about three

11

375 times larger than those of the SAM site (3.0×10^{11} liths m^{-2} yr^{-1}). The contribution of
376 intact coccospheres to the total coccolith export was low at both sites, with annual
377 coccosphere fluxes two orders of magnitude lower than coccolith fluxes at SOTS (3.5,
378 3.3 and 1.8×10^9 coccospheres m^{-2} yr^{-1} at 1000, 2000 and 3800 m, respectively) and SAM
379 (2.2×10^9 coccospheres m^{-2} yr^{-1}). Annualized CaCO$_3$ export was similar at both sites with
380 14.6, 16.2 and 17.1 g m^{-2} yr^{-1} at 1000, 2000 and 3800 m at the SOTS site and 13.9 g m^{-2}
381 yr^{-1} at the SAM sediment trap (1500 m).

382   Both coccolith and coccosphere fluxes displayed a marked seasonality that
383 followed the general trend of algal biomass accumulation in the surface waters at the
384 SOTS and SAM sites (Fig. 2). Coccolith fluxes at 1000 m started to increase in early
385 October and remained above the threshold of 1×10^9 coccoliths m^2 d^{-1} until mid-April
386 (Fig. 2). Three maxima were recorded during the period of high coccolith export:
387 October-early November 2011 (4×10^9 coccoliths m^2 d^{-1}), late December 2011 (9×10^9
388 coccoliths m^2 d^{-1}) and March 2012 (4×10^9 coccoliths m^2 d^{-1}). Coccolith fluxes of the
389 main coccolithophore species generally followed the similar seasonal pattern to that of
390 the total coccolith flux (Supplementary figure 1) and are not discussed further.
391 Coccolithophore fluxes registered by the 2000 and 3800 m sediment traps followed a
392 generally similar seasonal pattern to those of the shallower trap at the SOTS site (Fig. 2).
393 At SAM, coccolith fluxes exhibited a strong seasonality with peak fluxes in early January
394 2010 (up to 6×10^9 coccoliths m^2 d^{-1}) and a secondary peak in August 2010 (3×10^9
395 coccoliths m^2 d^{-1}). Coccosphere fluxes at both sites displayed maximum fluxes during the
396 austral summer and minima during winter; however maximum coccosphere export peaks
397 did not always match those of coccolith export (Fig. 2). The seasonality of total CaCO$_3$
398 followed a similar pattern to coccolith fluxes with peak values in the spring-summer and
399 minima during winter at both study sites.
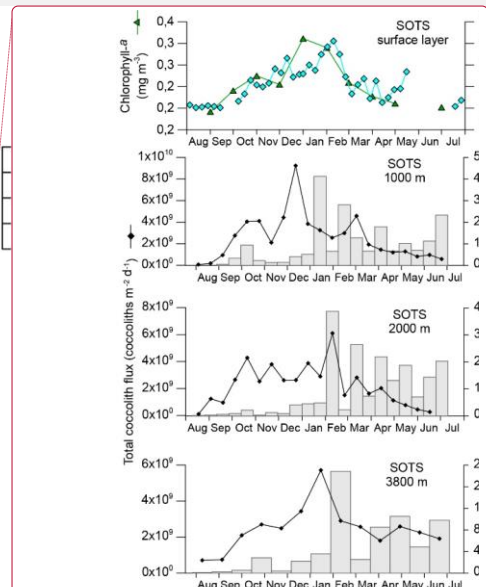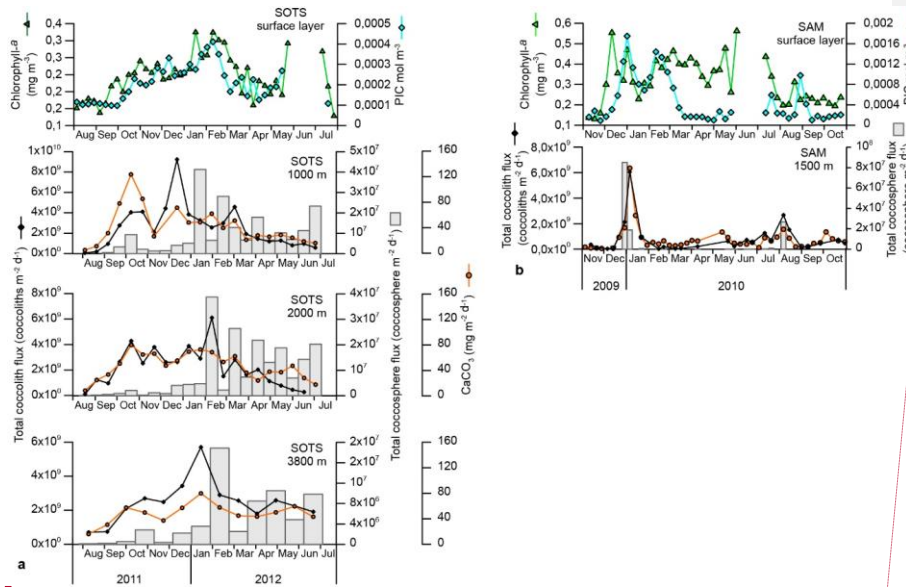
Figure 2: <u>Ocean-colour</u> satellite-derived chlorophyll-*a* and Particulate Inorganic Carbon (PIC) concentration in the surface layer and total <u>CaCO$_3$,</u> coccolith and coccosphere fluxes registered by the sediment traps at the SOTS (a) <u>and</u> SAM (b) <u>sites</u>.

3.2. Coccolithophore assemblage composition

Coccolith sinking assemblages were overwhelmingly dominated by *Emiliania huxleyi* for all sediment trap records analysed (Fig. 3a). At the SOTS site, the annualized flux-weighted relative contribution of *E. huxleyi* decreased slightly with depth, comprising 88% of the total coccolithophore assemblage at 1000 m, 82% at 2000 m and 80% at 3800 m. Secondary components of the coccolith sinking assemblage were *Calcidiscus leptoporus* (*sensu lato*) (6.8, 10.1 and 9.6% at 1000, 2000 and 3900 m, respectively), *Heliscosphaera carteri* (1.4, 2 and 1.3%) and small *Gephyrocapsa* spp. (< 3 µm) (1.4, 1.5 and 4.7%). Background concentrations (≤ 1%) of *Calciosolenia* spp., *Coccolithus pelagicus*, *Gephyrocapsa muellerae*, *Gephyrocapsa oceanica*, *Gephyrocapsa* spp. (> 3 µm), *Syracosphaera pulchra*, *Syracosphaera* spp., *Umbellosphaera tenuis* (*sensu lato*), and *Umbilicosphaera sibogae* were also registered. At the SAM site, *E. huxleyi* accounted for 83% of the annualized coccolith flux, with subordinate contributions of *C. leptoporus* (12.2%) and *Gephyrocapsa* spp. (< 3 µm) (1.5%). Background concentrations (< 1%) of *Calciosolenia* spp., *Coccolithus pelagicus,*

432 *G. oceanica*, *Gephyrocapsa muellerae, Gephyrocapsa* spp. (> 3 µm), *H. carteri,*

433 *Syracosphaera pulchra, Syracosphaera* spp., *U. sibogae* and *U. tenuis* were observed.
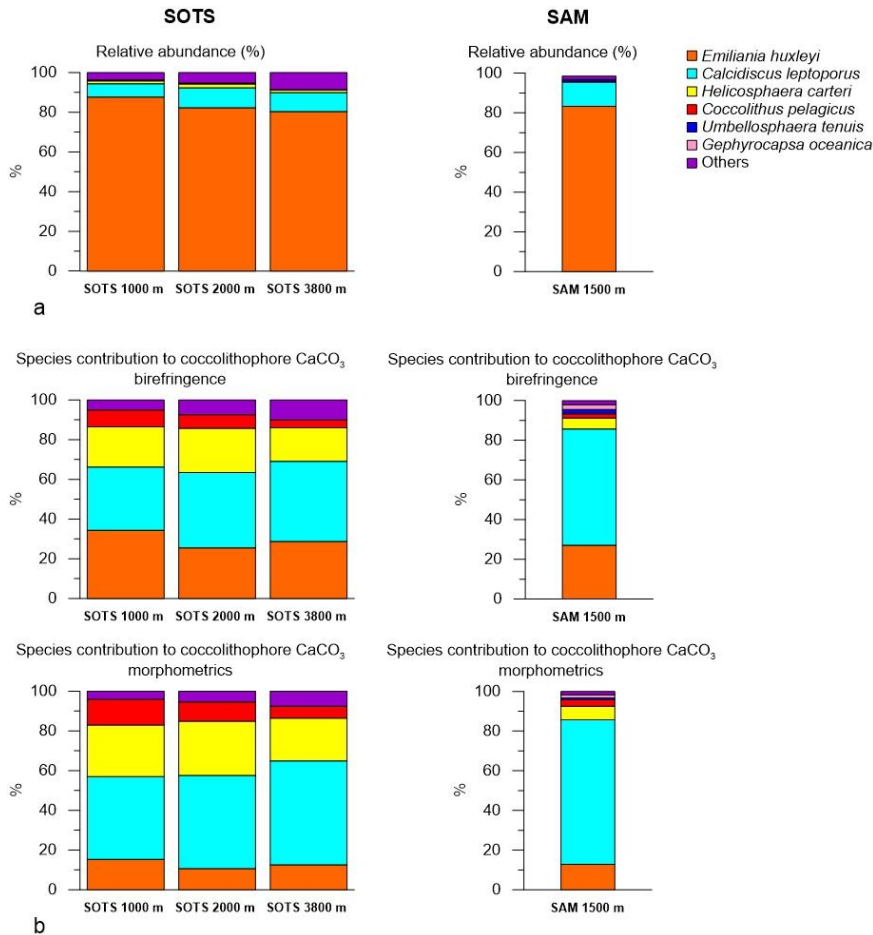
434 **Figure 3: a**. Annualized integrated relative abundance of the most important

435 coccolithophore species in the SOTS and SAM sediment trap records. **b**. Fractional

436 contribution of coccolithophore species to total coccolithophore CaCO$_3$ export in the
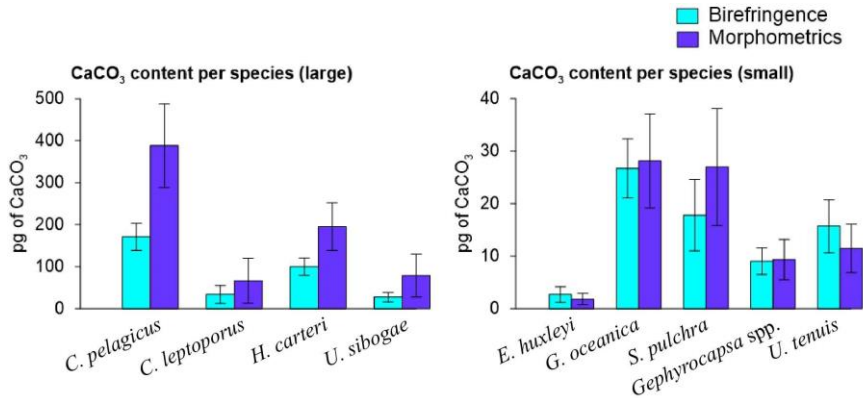
437 SOTS and SAM sediment traps.

438 **3.3 Calcite content per species**

439   Coccolith length and mass for all species measured using birefringence and

440 morphometric techniques are provided in Table 1. Overall, the average coccolith mass

441 estimates for the coccolithophore species at SOTS and SAM sites using both approaches

are within the range of values in the published literature. The Noelaerhabdaceae family members, *G. oceanica* and *Gephyrocapsa* spp., display almost identical mass values with both approaches (Fig. 4). In contrast, substantial discrepancies are identifiable for *C. pelagicus*, *C. leptoporus*, *H. carteri* and *U. sibogae,* for which coccolith mass estimates are about two-fold greater using morphometrics than with the birefringence approach. The range of annual contributions of coccolithophores to carbonate is illustrated in Figure 5.

Species and morphotypes	Type of measurement	n	Length (μm) Average	SD	Mass CaCO₃ (pg) Average	SD	k_s	Crystal units types	References
Calcidiscus leptoporus	Birefrigence	210	6.39	1.49	33.65	21.11	-	V and R	
	Morphometrics	210	6.39	1.49	66.23	53.28	0.080		1
	Literature estimates	-	4.3-9.6		22.6-125.2		0.061-0.105		1,2
Coccolithus pelagicus	Birefrigence	54	13.28	1.14	170.90	32.33	-	V and R	
	Morphometrics	54	13.28	1.14	387.96	99.64	0.060		1
	Literature estimates	-	8.5-13.5		99.5-398.6		0.051 - 0.060		1,2,3
Emiliania huxleyi	Birefrigence	12842	2.78	0.57	2.64	1.43	-	R	
	Morphometrics	12842	2.78	0.57	0.99-2.64 (1.81)*	0.60-1.60	0.015-0.04 (0.0275)*	(V-unit vestigial)	
E. huxleyi type A	Literature estimates	-	3-4		1.50 - 3.50		0.02		1,4,5
E. huxleyi type A o/c	Literature estimates	-	3.5		4.6		0.04		1
E. huxleyi type B/C	Literature estimates	-	1.8-5.5		0.3-3.5		0.015		5,6,7
E. huxleyi type B	Literature estimates	-	3.5-5		2.30 - 6.81		0.02		1,5
Gephyrocapsa oceanica	Birefrigence	51	5.87	0.62	26.70	5.64	-	R	
	Morphometrics	51	5.87	0.62	28.14	8.97	0.050	(V-unit vestigial)	
	Literature estimates	-	5-5.35		16.9-25.7		0.050-0.062		1,2
Gephyrocapsa spp.	Birefrigence	10	4.03	0.59	9.00	2.51	-	R	
	Morphometrics	10	4.03	0.59	9.33	3.84	0.050	(V-unit vestigial)	1
	Literature estimates	-	-	-	-		-		
Helicosphaera carteri	Birefrigence	64	11.20	1.12	100.10	20.34	-	V and R	
	Morphometrics	64	11.20	1.12	194.95	56.45	0.050		1
	Literature estimates	-	9.1-10		135-142.8		0.050-0.070		1,2
Syracosphaera pulchra	Birefrigence	81	6.77	1.09	17.77	6.80	-	V, R and T	
	Morphometrics	81	6.77	1.09	26.94	11.16	0.030		1
	Literature estimates	-	2.7-6		13.5-16.5		0.027-0.083		1,2,4
Umbellosphaera tenuis	Birefrigence	54	6.42	0.99	15.69	5.02	-	R	
	Morphometrics	54	6.42	0.99	11.45	4.61	0.015		1
	Literature estimates	-	5-6		8.7-23.9		0.015-0.071		1,2
Umbilicosphaera sibogae	Birefrigence	6	7.76	1.81	27.14	11.07	-	V and R	
	Morphometrics	6	7.76	1.81	78.93	51.38	0.055		1
	Literature estimates	-	4.1-6	-	16-35		0.055-0.086		1,2

Table 1: Coccolith mass estimates of the main coccolithophore species found at the SOTS and SAM sites using birefringence (C-Calcita) and morphometrics. Additionally, length and mass estimates from the literature are also listed for most species. References: (1) Young and Ziveri (2000), (2) Beaufort and Heussner (1999), (3) Samtleben and Bickert (1990), (4) Poulton et al. (2010), (5) Poulton et al. (2011), (6) Holligan et al. (2010) and (7) Charalampopoulou et al. (2016). * coccolith mass range obtained applying the minimum and maximum k_s values for *E. huxleyi* found in the literature (i.e. 0.015 and 0.04, respectively).

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Figure 4: Average and standard deviation of the coccolith mass estimates of the most important coccolithophore species captured by the SOTS and SAM sediment traps using birefringence (C-Calcita) and morphometric approaches. For *E. huxleyi,* the morphometric-based coccolith mass estimate was calculated by applying a mean shape factor constant (k_s) value estimated from the range of all the morphotypes found at the SAZ (i.e. $k_s = 0.0275$, Table 1).

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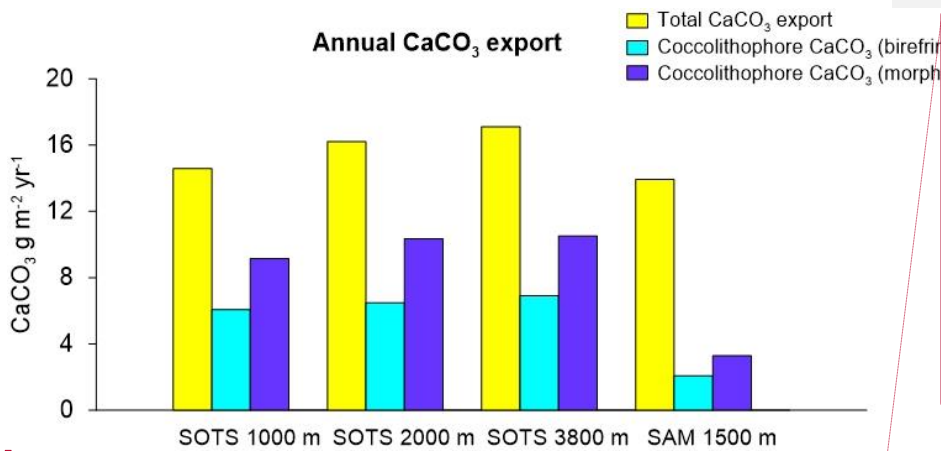
Figure 5: <u>Total annual $CaCO_3$ export (chemically determined) and fractional contribution of coccolithophores to $CaCO_3$</u> estimated using birefringence (C-Calcita) and morphometric approaches for the SOTS and SAM sites.

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4. Discussion

4.1 Coccolithophore phenology in the SAZ: satellite versus sediment trap records

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Total coccolith flux seasonality at the SOTS site shows good congruence with satellite-derived PIC in the surface layer, with both parameters suggesting enhanced coccolithophore productivity between October and March (austral mid-spring to early autumn; Fig. 2a). Interestingly, substantial coccosphere export (> 1 x 10^7 coccospheres $m^2\ d^{-1}$) does not occur until January indicating that coccolith and coccosphere export are not tightly coupled in the subantarctic waters south of Australia. Two different processes could be invoked to explain the mismatch between coccolith and coccosphere fluxes at this site. Firstly, *E. huxleyi*, the dominant coccolithophore species in the Southern Ocean, is able to produce coccoliths rapidly (up to three coccoliths per hour; Paasche, 1962; Balch et al., 1996) and shed the excess of coccoliths into the surrounding water under certain environmental conditions (Paasche, 2002). Although the coccolith shedding rate of *E. huxleyi* increases linearly with cellular growth rate (Fritz and Balch, 1996; Fritz, 1999), the tiny size and low weight of detached coccoliths allow them to remain in the upper water column long after cell numbers have begun to decline. It follows that high concentrations of detached coccoliths do not necessary imply a proportional abundance of coccospheres in the surface layer (Tyrrell and Merico, 2004; Poulton et al., 2013) or in the traps. Additionally, a substantial fraction of the coccospheres produced in the surface layer may experience substantial mechanical breakage by zooplankton before reaching the trap depths. Indeed, microzooplankton grazing pressure can remove up to 82% of primary production in mid-summer in the subantarctic waters south of Tasmania (Ebersbach et al., 2011) and about 60% of the daily coccolithophore growth in the North Atlantic (Mayers et al., 2019), therefore suggesting a strong top-down control on coccolithophore populations. Additionally, a polyacrylamide gel sediment traps study in the subantarctic waters south of Tasmania by Ebersbach et al. (2011) revealed that most of the particles exported out the mixed layer during the productive period occur in the form of faecal aggregates. Therefore, it is highly likely that: (i) the intensity of coccosphere export registered by the traps is influenced by grazing pressure in the surface layer, and (ii) that the impact of grazing on coccolithophores varies throughout the year (Calbet et al., 2008; Lawerence and Menden-Deuer, 2012; Quéguiner, 2013).

In contrast, seasonal variations in satellite-derived PIC concentration and coccolith fluxes at SAM show some discrepancies not observed at SOTS. While maximum PIC concentrations in the surface layer and coccolith and coccosphere fluxes co-occur in December and January (austral early to mid-summer), satellite-derived PIC suggests a secondary maximum in February-early-March not recorded by the trap (Fig.

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2b). One possibility is that the satellite secondary maximum is not coccoliths. The higher chlorophyll-a levels at the SAM site (Fig. 2) suggests that other phytoplankton groups, such as diatoms, are more abundant than in the subantarctic waters south of Tasmania. Empty and broken diatom valves have been suggested to display similar spectral characteristics than those of coccolithophore blooms (Broerse et al., 2003; Tyrrell and Merico, 2004; Winter et al., 2014). Therefore, the second peak in satellite-derived PIC could have been caused by a senescent diatom bloom. This hypothesis is likely since diatom blooms in the SAZ are known to develop early in the productive season (Rigual-Hernández et al., 2015b) and rapidly decay following the depletion of silicate and/or iron stocks in the surface layer (Lannuzel et al., 2011). However, no secondary late summer maximum was observed in biogenic silica fluxes in the SAM. Another possible explanation is a contribution to the satellite record from lithogenic material or storm-induced microbubble injection (Zhang et al., 2002). Fully resolving causes of mismatches between *in-situ* and satellite PIC estimates is not achievable for the SAM site (nor more broadly for the Southern Ocean; Trull et al., 2018).

A second difference between the SAM and SOTS sites is that maximum annual coccosphere export occurred one week earlier than maximum coccolith fluxes at SAM, (Fig. 2). The different seasonalities between the sites suggest that different export mechanisms may operate. The formation of rapidly sinking algal aggregates by diatoms is known to scavenge particles they have collided with and increase particle sinking (Alldredge and McGillivary, 1991; Passow and De La Rocha, 2006), thus the formation of such rapid-sinking aggregates could potentially facilitate the preservation of coccospheres early in the productive season at the SAM site. However, the lack of accompanying *in situ* information on plankton community structure in the study region precludes the assessment of these hypotheses.

Despite the uncertainties involved in our interpretations, the overall picture that emerges from our comparison of satellite and sediment trap flux data is that the duration of the coccolithophore bloom based on ocean-colour-based PIC concentrations most likely provides an over-estimation of the coccolithophore productive season. Our observations motivate caution in describing coccolithophore phenology solely based on satellite-derived PIC concentrations (e.g. Hopkins et al., 2015).

4.2 Magnitude and composition of subantarctic coccolithophore assemblages

548   Annual coccolith export across the major zonal systems of the Australian sector
549   of the Southern Ocean exhibits a clear latitudinal gradient, with maximum fluxes at the
550   SAZ (8.6 x 10^{11} liths m^{-2} yr^{-1}) and eight-fold lower fluxes in the polar waters of the AZ
551   (1.0 x 10^{11} liths m^{-2} yr^{-1}; Rigual Hernández et al., 2018). Coccolithophore species
552   occurrence documented by our subantarctic sediments traps are consistent with previous
553   reports on coccolithophore assemblage compositions in the surface layer (Findlay and
554   Giraudeau, 2000; Saavedra-Pellitero et al., 2014; Malinverno et al., 2015; Chang and
555   Northcote, 2016) and sediments (Findlay and Giraudeau, 2000; Saavedra-Pellitero and
556   Baumann, 2015) and are more diverse than those found in the AZ (Rigual Hernández et
557   al., 2018). The southward decline in coccolithophore abundance and diversity is most
558   likely due to the decrease in sea-surface temperature (SST) and light availability moving
559   poleward (Charalampopoulou et al., 2016; Trull et al., 2018). In particular, the close
560   relationship between temperature and growth rates has been demonstrated in a range of
561   coccolithophore species and strains (Buitenhuis et al., 2008), and seems to be a critical,
562   if not the most important, control on the biogeographical distribution of coccolithophore
563   species in the Southern Ocean (Trull et al., 2018). This pronounced latitudinal change in
564   coccolithophore assemblage composition contrasts with the little longitudinal variability
565   between the subantarctic SOTS and SAM sites (Fig. 3). These observations underscore
566   the role of circumpolar fronts as natural physical barriers for plankton species distribution
567   in the Southern Ocean (Medlin et al., 1994; Boyd, 2002; Cook et al., 2013).

568   Notably, the rare occurrence of the cold-water species *Coccolithus pelagicus* at
569   the SOTS and SAM sites contrasts with the high contribution of *C. pelagicus* to the living
570   coccolithophore communities in the subpolar and polar waters of the North Atlantic and
571   North Pacific oceans, where it is often the second most abundant species after *E. huxleyi*
572   (McIntyre and Bé, 1967; Baumann et al., 2000; Broerse et al., 2000a; Broerse et al.,
573   2000b; Ziveri et al., 2000). This important difference in species composition between
574   Northern and Southern hemisphere subpolar ecosystems could have important
575   implications in the calibration of the satellite PIC signal in the Southern Ocean. Previous
576   research in the Southern Ocean comparing satellite and shipboard observations identified
577   a substantial over-estimation of coccolithophore PIC in the Southern Ocean waters by
578   satellite ocean-colour-based PIC algorithms (Holligan et al., 2010; Trull et al., 2018).
579   Since satellite reflectance observations are mainly calibrated against Northern
580   Hemisphere PIC results (Balch et al., 2011; Balch et al., 2016), the lower the calcite
581   content of dominant *E. huxleyi* morphotypes (B/C) in the Southern Ocean compared to

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587 their northern hemispheric counterparts has been suggested as a possible factor causing
588 the over-estimation of PIC concentrations in the Southern Ocean. Following this
589 reasoning, we speculate that differences in other components of the coccolithophore
590 assemblages, and particularly, differences in *C. pelagicus* numbers, could contribute to
591 the over-estimation of PIC concentrations by the satellite PIC algorithm in the Southern
592 Ocean. Indeed, the scaling of reflectance (in satellite images) to PIC (in ocean) is very
593 dependent on coccolith area:mass ratios (Gordon and Du, 2001; Balch et al., 2005a).
594 *Coccolithus pelagicus* has remarkably heavier and thicker coccoliths (100-400 pg per
595 coccolith; Table 1) than *E. huxleyi* (~3 pg per coccolith), i.e. about 100 times heavier.
596 However, the average coccolith area of *C. pelagicus* is only about ten times greater than
597 that of *E. huxleyi*. Thus, this lack of proportional relationship between area and mass
598 between these species should be taken into consideration when calibrating the satellite
599 signals of coccolithophore-related PIC in the Southern Ocean. However, it should be
600 noted that this is only one possible factor contributing to the overestimation of PIC
601 concentrations in Southern Ocean waters. Other factors such as the presence of
602 microbubbles -- that are a source of enhanced reflectance -- must also play an important
603 role (Balch et al., 2011).

604

605 **4.3 Coccolith calcite content of subantarctic coccolithophore species**

606   Multiple methodological biases associated with each of the methods used for
607 estimating coccolith calcite content (i.e. birefringence, morphometrics) could be invoked
608 to explain the different estimates observed for some of the species (see Young and Ziveri,
609 2000; Fuertes et al., 2014 and references therein). However, the fact that these
610 discrepancies vary greatly across species suggests that the composition of the crystal-
611 units of the coccoliths could be the most important factor causing these differences. All
612 the heterococcoliths of the species analysed are mainly composed of either V- or R-
613 calcite crystal units or a combination of both (Young et al., 2003; Table 1). R units are
614 characterized by sub-radial c-axes that are reasonably well measured by the birefringence
615 technique, but, the almost vertical optical axes of the V units (Young, 1992; Young et al.,
616 1999) make the same thickness less birefringent (Fuertes et al., 2014). Thus, it is likely
617 that differences in the birefringence properties of the R and V units could be responsible
618 for the different estimates provided by the two approaches. This is supported by our
619 results which show coccolith mass estimates of those species composed of R units, such

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Eliminado: we speculate that differences in the coccolithophore assemblage composition, and particularly, differences in *C. pelagicus* numbers, could contribute to the over-estimation of PIC concentrations by the satellite PIC algorithm in the Southern Ocean

as *G. oceanica* and *Gephyrocapsa* spp. exhibit almost identical values with both techniques (Table 1). In contrast, those species with coccoliths composed by a combination of R and V units, such as *C. pelagicus*, *C. leptoporus*, *H. carteri* and *U. sibogae,* display divergent mass estimates between approaches. The case of *E. huxleyi* is more complex due to the large intraspecific genetic variability that results in substantial differences in the profile and degree of calcification between specimens (Young and Ziveri, 2000). Our birefringence mass estimate for *E. huxleyi* (2.67 ± 1.49 pg) is less than one picogram lower than the mean range value calculated with the morphometric technique (i.e. 1.81 ± 1.10 pg with an average k_s value of all the morphotypes found at the SAZ, i.e. $k_s = 0.0275$), but identical to the maximum (2.64 ± 1.60 pg; using $k_s = 0.04$). These results suggest a reasonably good consistency between techniques for *E. huxleyi*.

Taking into consideration all the above, it is likely that the coccolith mass of some species is underestimated by the birefringence technique, and therefore, the fractional contribution of coccolithophores to total PIC using this approach should be taken as a conservative estimate. Since both methods for estimating calcite content have inherent uncertainties, the range of values provided by both techniques is used here as an approximation of the fractional contribution of coccolithophores to total annual $CaCO_3$ export to the deep sea in the Australian and New Zealand sectors of the SAZ.

4.4 Contribution of coccolithophores to carbonate export in the Australian-New Zealand sectors of the Southern Ocean

The magnitude of the total PIC export in the subantarctic waters was similar between the SOTS and SAM sites (range 14-17 g m^{-2} yr^{-1}), and thus slightly above the global average (11 g m^{-2} yr^{-1}; Honjo et al., 2008). Our estimates indicate that coccolithophores are major contributors to $CaCO_3$ export in the Australian and New Zealand waters of the SAZ, accounting for 40-60% and 15-25% of the annual $CaCO_3$ export, respectively (Fig. 5). Heterotrophic calcifiers, mainly planktonic foraminifera (Salter et al., 2014), must therefore account for the remainder of the annual $CaCO_3$ export at both sites. Previous work on foraminifera fluxes in our study regions allows an approximate estimate of the contribution of foraminifera to total $CaCO_3$ flux that can be used to assess the validity of our estimates. Combining counts of foraminifera shells (King and Howard, 2003) with estimates of their average shell weights (20-40 µg per shell depending on size; Moy et al., 2009) suggests contributions of 1/3 to 2/3 of planktonic foraminifera to the total $CaCO_3$ flux in the Australian SAZ (Trull et al., 2018).

658 In the subantarctic waters south of New Zealand, Northcote and Neil (2005) estimated
659 that planktonic foraminifera accounted for about 78-97% of the total $CaCO_3$. Thus,
660 estimations of the contribution of heterotrophic calcifiers to total carbonate in both study
661 regions are in reasonable agreement with our coccolithophore $CaCO_3$ estimates at both
662 sites. The lower contribution of coccolithophores to $CaCO_3$ export at the SAM site in
663 comparison with that of SOTS may be explained by differences in the ecosystem structure
664 between sites. Algal biomass accumulation in the surface waters of the SAM region
665 (average chlorophyll-*a* concentration between 2002 and 2018 is 0.31 mg m^{-3}) is
666 substantially higher than that at SOTS (0.23 mg m^{-3}). We speculate that the higher
667 abundance of non-calcareous phytoplankton (e.g. diatoms) in the subantarctic waters
668 south of New Zealand could simultaneously reduce coccolithophore biomass through
669 resource competition (Quéré et al., 2005; Sinha et al., 2010) while stimulating
670 foraminifera growth (Schiebel et al., 2017). The combination of both factors could be
671 responsible for the lower coccolithophore productivity at the SAM site despite similar
672 total $CaCO_3$ export. Assuming that both the SOTS and SAM sites can be considered
673 representative of a broad longitudinal swath of the SAZ south of Australia and New
674 Zealand (ca. 1% of areal extent of the global ocean), the coccolithophore $CaCO_3$ export
675 in these two regions together account for approximately 0.4 Tmol C_{inorg} yr^{-1}. This value
676 represents approximately 1% of the global annual PIC export to the deep ocean (Honjo et
677 al., 2008) and underscores the notion that the high nutrient low-chlorophyll waters of the
678 circumpolar SAZ should not be taken as indicative of low biological activity or export.

679   Our results indicate that although *E. huxleyi* overwhelmingly dominates the
680 coccolithophore sinking assemblages at both study sites, other species with lower relative
681 contribution but substantially heavier coccoliths are more important contributors to the
682 annual coccolithophore $CaCO_3$ export budget (Fig. 3). Particularly relevant is the case of
683 *C. leptoporus* that despite its relatively low abundance (~ 10% of the annual assemblage
684 at both sites; Fig. 3), it accounts for between 30-50% and 60-70% of the annual
685 coccolithophore-$CaCO_3$ export at the SOTS and SAM sites, respectively (Fig. 3).
686 Similarly, other species with heavy coccoliths, such as *H. carteri* and *C. pelagicus*, are
687 important contributors to the annual coccolithophore PIC export to the deep sea (up to
688 ~30% and ~10% of the annual coccolithophore PIC, respectively) despite their low annual
689 relative abundance (<2% at both sites) (Fig. 3). These results serve as an important
690 reminder that it is often not the most abundant species, but rather the largest

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692 coccolithophore species that account for the greatest contribution to coccolithophore
693 CaCO$_3$ production and export (Young and Ziveri, 2000; Baumann et al., 2004; Daniels et
694 al., 2016).

695   The important contribution made by the coccolithophore community in setting the
696 magnitude of carbonate production and export to the deep sea is evidenced when we
697 compare the coccolith and total CaCO$_3$ fluxes of the SOTS sediment trap with those
698 deployed in the AZ along the 140°E meridian (Fig. 1). Although both total and
699 coccolithophore CaCO$_3$ export decrease with increasing latitude these changes are largely
700 uneven. While total CaCO$_3$ decreases two-fold from the SAZ to the AZ, coccolithophore
701 CaCO$_3$ export decreases 28-fold (Supplement Figure 2). This lack of proportional
702 latitudinal change can be attributed to two main factors. First, subantarctic
703 coccolithophore populations are diverse and relatively rich in species with large and
704 heavy coccoliths such as *C. leptoporus* or *H. carteri* that account for a large fraction of
705 the annual carbonate production and export. South of the PF, assemblages become
706 monospecific, or nearly monospecific, dominated by the small and relatively lightly
707 calcified *E. huxleyi*. Second, latitudinal variations in the abundance of heterotrophic
708 calcifiers (mainly foraminifera but also pteropods) must play a major role in modulating
709 the observed variations in CaCO$_3$ export. In particular, our data suggests that the
710 fractional contribution of heterotrophic calcifiers to CaCO$_3$ production increases from
711 ~40-60 % in the Australian SAZ to up to 95% in the AZ (Rigual Hernández et al., 2018).
712 This pattern is consistent with previous shipboard and sediment trap studies that reported
713 higher abundances of planktonic foraminifera at the PFZ and AZ compared to that of the
714 SAZ in the Australian sector (King and Howard, 2003; Trull et al., 2018). Controls on the
715 biogeographic distribution of foraminifera species are complex and beyond the scope of
716 this paper, however, we provide a few observations. Both temperature and diet are critical
717 factors controlling the spatial distribution of planktonic foraminifera species. In
718 particular, the lower temperatures south of the SAF seem to favour the development of
719 *Neogloboquadrina pachyderma* sin. and *Turborotalita quinqueloba* as indicated by the
720 high abundance of these species in the PFZ (> 80% of the annual integrated flux for both
721 species together; King and Howard, 2003). Additionally, the dramatically different algal
722 communities dwelling in each zonal system may also play a role in planktonic
723 foraminifera species distributions. In particular, diatoms can account for a major part of
724 the diet of some foraminifera species, including *N. pachyderma* (Schiebel and Hemleben,

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725 2017). Therefore, it is likely that the preferential grazing on diatoms of some foraminifera
726 species may play an important role in the increase of foraminifera $CaCO_3$ production
727 moving poleward.

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729 **4.5 Future predictions of coccolithophore community response to environmental**
730 **change in the subantarctic zone**

731 The response of *E. huxleyi* to environmental change has been extensively studied
732 in laboratory experiments (Meyer and Riebesell, 2015; Müller et al., 2015; Feng et al.,
733 2017) and the available information is sufficient to propose possible changes of its niche
734 and calcification in the Southern Ocean, as discussed in detail in Trull et al. (2018) and
735 Krumhardt et al. (2017). Due to the ubiquity and abundance of *E. huxleyi*, the
736 ecophysiology of this species is often regarded as typical of all coccolithophores.
737 However, *E. huxleyi* is rather different from most other coccolithophore species in that its
738 physiological adaptations place it in the upper limit of the *r-K* ecological gradient of these
739 organisms (i.e. an opportunistic species), while most of the other species are located at
740 the opposite end of the spectrum (i.e. conservative or K-selected species) (Probert and
741 Houdan, 2004). Our results demonstrate that *E. huxleyi* plays an important, but not
742 dominant role in $CaCO_3$ export, with other species such as *C. leptoporus, H. carteri* or *C.*
743 *pelagicus* making a larger contribution to the annual $CaCO_3$ export in the SAZ (Fig. 3).
744 Therefore, it is of critical importance to evaluate how these other biogeochemically
745 important coccolithophore species will respond to projected climate-induced changes in
746 the Southern Ocean. Here, we now assess the response of large coccolithophore species
747 to projected changes in temperature and carbonate chemistry, that have been highlighted
748 among the most important environmental stressors expected to impact Southern Ocean
749 coccolithophore physiological rates (Müller et al., 2015; Charalampopoulou et al., 2016;
750 Feng et al., 2017; Trull et al., 2018).

751 The Southern Ocean is warming rapidly (Gille, 2002; Böning et al., 2008), largely
752 due to the southward migration of the ACC fronts (Sokolov and Rintoul, 2009). Only
753 between 1992 and 2007 the position of Southern Ocean fronts shifted by approximately
754 60 km to the south (Sokolov and Rintoul, 2009) and this trend may continue throughout
755 the next century. Therefore, it is likely that any further southward migration of ACC
756 fronts will be coupled with an expansion of subantarctic coccolithophore species towards
757 higher latitudes. The poleward expansion of *E. huxleyi* geographic range has already been

suggested in the Southern Ocean (Cubillos et al., 2007; Winter et al., 2014; Charalampopoulou et al., 2016) and it also appears to be occurring in the North Atlantic (Rivero-Calle et al., 2015; Neukermans et al., 2018). Given the important contribution of large subantarctic coccolithophore species to $CaCO_3$ export, the expansion of their ecological niche could result in a substantial increase in $CaCO_3$ production and export in the Southern Ocean. However, this may not be the future scenario for the SAZ southeast on New Zealand, where bathymetry strongly controls the location of ocean fronts (Fernandez et al., 2014; Chiswell et al., 2015). If the fronts are bathymetrically 'locked', then the SAZ will not expand in areal extent, although the region is still predicted to undergo significant physical, biogeochemical and biological changes (Law et al., 2017) that will have likely flow-on effects on coccolithophore productivity and export (Deppeler and Davidson, 2017).

The available carbonate chemistry manipulation experiments with *C. leptoporus* have come to different conclusions. While some studies identified an increase in coccolith malformations with increasing CO_2 concentrations (Langer et al., 2006; Langer and Bode, 2011; Diner et al., 2015), another study (Fiorini et al., 2011) reported no changes in the calcification of *C. leptoporus* at elevated pCO_2. Interestingly, *C. leptoporus* did not experience changes in its photosynthesis rates over the tested CO_2 range in any of the aforementioned studies. The most likely explanation for the different results between the studies is a strain-specific variable responses to changing carbonate chemistry (Diner et al., 2015). Strain-specific variability in response to changing carbonate chemistry has been previously reported in other coccolithophores, such as *E. huxleyi* (Langer et al., 2009; Müller et al., 2015), and therefore it is likely that this also occurs in other species. Given the fact that Southern Ocean fronts act as barriers for species distributions and gene flows (Medlin et al., 1994; Patarnello et al., 1996; Thornhill et al., 2008; Cook et al., 2013), it is possible that the subantarctic *C. leptoporus* populations exhibit a different ecophysiology than those used in the above mentioned laboratory experiments. Prediction of the responses of *H. carteri* and *C. pelagicus* is even more challenging due to the lack of experiments testing the response of these species to changing seawater carbonate chemistry. The only available insights in the response of one of these species to ocean acidification are found in the fossil record. Both Gibbs et al. (2013) and O'Dea et al. (2014) reconstructed the evolution of *C. pelagicus* populations during the Palaeocene-Eocene Thermal Maximum (PETM), a period arguably regarded as the best geological approximation of the present rapid rise in atmospheric CO_2 levels and temperatures.

794 These studies concluded that *C. pelagicus* most likely reduced its growth rates and
795 calcification during this period. This limited number of studies suggest that the ongoing
796 ocean acidification in the Southern Ocean could potentially have a negative impact on the
797 physiological rates of *C. leptoporus* and *C. pelagicus* while the effect on *H. carteri* is
798 unknown. Physiological response experiments (e.g. Müller et al., 2015) with Southern
799 Ocean strains of *C. leptoporus*, *H. carteri* and *C. pelagicus* are, therefore, urgently needed
800 to be able to quantify the effect of projected changes in oceanic conditions in the SAZ on
801 their physiological rates and consequent effects on carbon cycling in the Southern Ocean.

802 Our synthesis suggests opposing influence of environmental stressors on
803 subantarctic coccolithophore populations. Poleward migration of fronts will likely
804 increase coccolithophore $CaCO_3$ production in the Southern Ocean, while changes in
805 carbonate chemistry speciation will reduce growth rates of subantarctic coccolithophores.
806 It seems possible that coccolithophores will initially expand southward as waters warm
807 and fronts migrate, but then eventually diminish as acidification overwhelms those
808 changes.

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826 **Author contributions**

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Competing interests

The authors declare no competing interests.

Data Availability

Morphometric data of major coccolithophore species generated during the current study are listed in Table 1, while species relative abundance and species fluxes (plotted in Supplement Figure 1) can be accessed in the following link:

https://data.aad.gov.au/metadata/records/Coccolithophore_Fluxes_SAZ_2009-2012.

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