All referee comments are in bold and our answers in normal font. Changes in the manuscript text are kept in italics.

Referee #2

This manuscript contributes important empirical evidence and debate on the influence that ocean acidification may have on biogeochemistry in the oceans. Iron availability is episodic in the NE Pacific and plays an important part in determining pelagic community composition and levels of productivity. Investigating how an increase in pCO2/decrease in pH influences the response to episodic iron availability in natural oceanic communities is ambitious and unsurprisingly, not altogether easy to interpret. Nonetheless, the authors have designed an effective and achievable experiment, applied a suite of relevant measurements and provided, in general, a balanced interpretation of the results. Several points could be clarified to improve the manuscript:

1. Title: The title fails to capture the essence of the study, in my view. It puts a large emphasis on the influence of dust additions but the manuscript is presented very much as an ocean acidification+dust experiment. It also does not capture the impact on community composition, which is a much larger component of the paper than the DMS response.

Reply: We thank the reviewer for pointing that out. The title has been reworded to:
Impact of ocean acidification on phytoplankton assemblage, growth and DMS production following Fe-Dust additions in the NE Pacific HNLC waters

2. Iron treatments and bioavailability: The application of two different versions of iron addition is useful and adds a great deal to the interest of the paper. However, this is not reflected in the stated objective of the research (L12286, L12+) and should be (1). How do differences in the form of iron used for previous comparable experiments influence those results (Introduction, L12285, L3+)? (2) Although the reader is directed to Nishikawa et al (2000) and Hwang and Ro (2006) for more information on the specific dust added, it would be useful to present more details on the dust in this context. This should include total iron content and possibly the form of the iron present in the dust (3). If DFe is not a good measure of the bioavailability of iron (Section 4.3) what should be measured? (4) Please discuss why the Fe contained in the dust is more available than the added FeSO4 (5).

Reply: (1) A reference to $FeSO_4$ was added to the objective.

New paragraph: *The objective of this study was to determine how a decrease in pH by 0.2 units could influence the impact of Fe delivered as $FeSO_4$ or Asian dust on the growth and taxonomic composition of the phytoplankton assemblage of the Fe-limited northeast subarctic Pacific in summer, and to explore how pH-induced changes could affect the production of the climate-active gas DMS.*

(2) To our knowledge, this is the first study using a natural form of iron, i.e. dust, in the context of a HNLC region. Studies intercomparison is not possible. A comparable protocol was used in a

study conducted in Atlantic where the addition of Sahara dust stimulated nitrogen fixation (Mills et al., 2004). In this study, the addition of FeSO4 and dust had similar effect on growth. But this last study is not directly relevant to our study.

(3) As requested, more information has been added on CJ-2 dust (see reply to referee #1).

(4) This question does not have a simple answer, since iron bioavailability doesn't have a universally accepted definition and methods used to quantify it also vary amongst laboratories. However, in a previous study, we showed that TDFe concentrations had a better predictive capacity than DFe values. We are referring to this previous work in the revised version of the manuscript.

New sentence: *Similar conclusions were reached in a previous study (Mélançon et al., 2014), where TDFe was also measured and shown to be a better indicator of Fe bioavailability than DFe.*

(5) Unfortunately, we can only speculate on that. A new sentence has been added in section 4.3 to explain the possibilities.

New sentence: *We can only speculate as to why Fe contained in dust stimulated more growth than $FeSO_4$. Part of the answer may be related to the speed at which the Fe is released from dust, a relatively low release rate allowing a more efficient utilization of Fe. In comparison, and as can be seen in Fig. 1, $FeSO_4$ is rapidly utilized and probably scavenged by particles and ligands following fertilization.*

3. Phytoplankton taxonomy and pigments (Section 2.4.3): The study uses HPLC based pigment analysis and Chemtax to assign class-specific contributions to total chlorophyll. This requires more explanation. In particular, how is the contribution of diatoms differentiated from that of haptophytes or pelagophytes? As Table 3 illustrates, these classes share several of the biomarker pigments and differentiating them is far from exact. At the last, the authors should make clear the uncertainty in the class-specific chlorophyll assignment and take it into account in their conclusions that increased pCO2 increased the fitness of diatoms over other taxa.

Reply: Fucoxanthin is the main biomarker of diatoms but, as shown in Table 3 and pointed out by the reviewer, fucox is also present in haptophytes and pelagophytes. In contrast, 19'-butanoyloxyfucoxanthin (but) is present in haptophytes and pelagophytes but not in diatoms. Thus, an increase in the concentration in "fucox" that is not accompanied by an increase in "but" can be used to differentiate the contribution of diatoms from that of haptophytes or pelagophytes. The method has been described in more detail (see reply to referee #1, and modifications in the text), however, we do not feel it is necessary to present raw pigment data. Our result do not show increased fitness of diatoms over other taxa linked to increased pCO_2 as there is no significant difference in the concentration of diatoms between acidified and non-acidified treatments. The increase in diatom biomass is more likely caused by iron fertilization.

4. Increased respiration is provided as one explanation for why increased 14C assimilation is not reflected in increased biomass in Control+Acid (P12298, L23+) and Dust+Acid (P12301, L19+). However, using 24h 14C incubations to determine carbon

assimilation is generally thought to measure something closer to net production than gross production; meaning that any increase in the rate of respiration would be captured by that measurement.

Reply: Good point. This explanation was deleted from the list.

5. DMSP and DMS. Given the generally high DMSP content of dinoflagellates, it is puzzling that the almost immediate loss of dinoflagellates is not reflected in the trends in DMSP? What/who were the main contributors to the DMSP pools? Are the high initial DMS:DMSP ratios (roughly 1:4) a product of the experimental set-up, i.e. cell disruption/ negative impact on dinoflagellates, or were in situ DMS concentrations similar at the time the water was sampled?

Reply: We cannot ascertain how fast the loss of dinoflagellates took place, since measurements were made only at T0 and T4. The DMSPt (which also includes dissolved DMSP) could decrease as dinoflagellates lysed and DMSPd got consumed by bacteria. It is likely that dinoflagellates and haptophytes were the main contributors to the DMSP pool, and the DMSP decrease was caused by the loss of dinoflagellates.

The DMS concentration is not unusually high. The northeast subarctic Pacific is known for very high DMS concentrations and high DMS:DMSP ratios. In situ DMS concentration at the same station and depth 17h prior to our water collection was ~15 nmol L^{-1}, a value relatively close to the value of 10 nmol L^{-1} measured at the beginning of the incubation. A note on the usually high concentration of DMS in this region has been added in the section 4.2 of the revised manuscript.

New sentence: *DMS concentrations were high, but usual for this region in the summer (Wong et al., 2005).*

6. Iron uptake rate. Figure 4C. I am not sure this shows Fe uptake rate. This was measured as the incorporation of added 55FeDFB. A clear explanation (Section 2.4.4) of how (and why) Fe uptake rates are calculated from the assimilation of the 55FeDFB complex is required.

Reply: We have amended the paragraph to provide an explanation for examining Fe uptake from DFB. Since uptake of Fe from DFB makes up a small portion of the manuscript, and is not the primary focus of it, we refer to previously published work that provides a thorough explanation for how the Fe uptake rates are calculated (in section 2.4.4; Maldonado and Price 1999; Semeniuk et al. 2009). However, we have provided a few more details to clarify how the rates were calculated.

Revised section 2.4.4

The siderophore desferrioxamine B (DFB) has been used as a model ligand for studying the bioavailability of strongly organically complexed Fe in seawater (e.g. Maldonado and Price 1999; Hutchins et al. 1999; Shi et al. 2010). Iron uptake from DFB occurs through a high affinity Fe transport system, and can reflect the Fe nutritional status of laboratory phytoplankton strains

(Maldonado and Price 1999; Maldonado et al. 2006) and natural phytoplankton assemblages (Semeniuk et al. 2009; Taylor et al. 2013; Semeniuk et al. in press). Thus, we examined whether uptake rates of Fe complexed to DFB varied among our treatments with varying Fe bioavailability.

Uptake of 55Fe bound to DFB was performed as previously described (Maldonado and Price 1999; Semeniuk et al. 2009). The 0.5 nM 55Fe (Perkin Elmer) was complexed with 5% excess DFB in pH 3.5 Milli Q for 30 minutes (Maldonado and Price, 1999). The resulting 55FeDFB complex was subsequently equilibrated in 0.22 µm filtered seawater for 2 h. Approximately 250 mL of seawater was subsampled from each incubation bag into acid-cleaned polycarbonate bottles. Just before dawn, the equilibrated 55FeDFB complex and 10 µCi of H14CO3- (Perkin Elmer) were added to each 250 mL bottle. From each assay bottle, 1 mL of sample was taken in order to determine the initial total added activities of 55Fe and 14C. To prevent inorganic 14C from off-gassing in the initial activity sample vial, 500 µL of 5 M NaOH was added.

After 24 h, the content of each bottle was gently filtered onto a 47 mm diameter, 1 µm porosity polycarbonate filter (AMD) under low vacuum pressure (≤70 mm Hg). Just before going dry, the filters were immersed in 5 mL of Titanium-ethylenediaminetetraacetic acid (Ti-EDTA) wash to remove extracellular Fe (Hudson and Morel, 1989). The filters were then rinsed with 10 mL of filtered seawater to remove any loosely associated tracer. Filters were placed into 7 mL borosilicate scintillation vials, immersed in 5 mL Scintisafe 50% scintillation cocktail, and conserved in the dark until analysis on a Beckman LS65005514 scintillation counter.

Volumetric Fe uptake and C-fixation rates were calculated as described elsewhere (Maldonado and Price 1999; Semeniuk et al. 2009). Previous work with phytoplankton assemblages along Line P has demonstrated that uptake of Fe from DFB by natural phytoplankton communities is linear over 24 h (Maldonado and Price 1999). Thus, Fe uptake rates were calculated assuming the accumulation of 55Fe by cells was linear during the assay. In order to compare Fe uptake rates among treatments, volumetric rates were normalized to the amount of C fixed during the assay. The 14C uptake rates normalised to chl a is used here as an indicator of the growth status of the autotrophic assemblage.

7. In general, the table and figure legends would benefit from a greater level of explanation.

Reply: Sample size was added as requested by reviewer 1, and more information was added to the legendsof tables and figures (see below).

Here are the modified table/figure titles:

Table 1: Description of Fe/dust additions and acidification status of experimental conditions for each treatment. All treatments were conducted in triplicate.

Table 3: Biomarker pigment initial ratio matrix for CHEMTAX: Chl a ratios for eight algal groups.

Figure 1: Average concentration of DFe in each treatment during the incubations measured at T0, T2 and T4. (a) Control and Control+Acid. (b) Fe and Fe+Acid. (c) Dust and Dust+Acid. Error bars indicate standard deviations. n = 3 except for Acid, T0, T2, Dust+Acid, T0, and Control (all times) where n = 2.

Figure 2: Average concentration of chl a (left axis) during the incubations and POC at T4 (right axis) in each treatment. (a) Control and Control+Acid. (b) Fe and Fe+Acid. (c) Dust and Dust+Acid. Error bars indicate standard deviations. Dashed line indicates POC concentration at T0. Chl a: n = 3 except for Acid, T0, Dust+Acid, T4, Fe+Acid, T0 and Control (all times) where n=2 because of missing/unreliable data or contamination (Control 1). POC: n=3 except Control where n=2.

Figure 3: Average chl a concentration (µg L-1) attributable to each of the measured groups of phytoplankton initially (T0: white bar) and for each of the treatments after 4 d (T4) of incubation (Control, Control+Acid, Fe, Fe+Acid, Dust, Dust+Acid; gray bars). (a) Diatoms. (b) Dinoflagellates. (c) Haptophytes. (d) Pelagophytes. (e) Chlorophytes. (f) Cyanobacteria. Error bars indicate standard deviations. n = 2 except for T0 (all groups but dinoflagellates) where n = 3 and Acid, diatoms, Control, haptophytes and pélagophytes, Dust+Acid, cyanobacteria and Fe+Acid, haptophytes and cyanobacteria where n = 1.

Figure 4: Average (a) Absolute C assimilation rates. (b) C assimilation rates normalized to chl a concentration at T4 and (c) Fe uptake rates normalized to chl a concentration at T4. Error bars indicate standard deviations. Absolute C assimilation rates: n = 3 except Control and Acid where n = 2, C assim norm to chl a: n = 3 except Control, Acid and Dust+Acid where n=2, Fe uptake rates normalized to chl a: n = 3 except Control and Dust+Acid where n = 2.

Figure 5: DMSPt (a, b and c) and DMS (d, e and f) concentrations (nmol L-1) in the Control and Control+Acid treatments (a and d), the Fe and Fe+Acid treatments (b and e), and the Dust and Dust+Acid treatments (c and f). Error bars indicate standard deviations.. n = 3 except Control where n = 2.

Minor points:
P12287, L19, CO2SYS needs to be consistent.

Reply: corrected to CO2SYS.

P12291, L16. What size of filter was used?

Reply: 47 mm diameter. The information has been added to the text.

P12294, L3 etc. the levels of precision, reflected in the number of decimal places, should be consistent.

Reply: Done.

P12295, L10. Maybe refer to the Control in this sentence.

Reply: Done.

New references cited

Hutchins, D. A., V. M. Franck, M. A. Brzezinski, K. W. Bruland (1999), Inducing phytoplankton iron limitation in iron-replete coastal waters with a strong chelating ligand, Limnol. Oceanogr., 44, 1009–1018.

Semeniuk, D.M., Taylor, R.L., Bundy, R.M., Johnson, W.K., Cullen, J.T., Robert, M., Barbeau, K.A., and M.T. Maldonado (in press), Iron–copper interactions in iron-limited phytoplankton in the northeast subarctic Pacific Ocean, Limnol. Oceanogr. doi: 10.1002/lno.10210.

Wong, C. S., Wong, S. E., Richardson, W. A., Smith, G. E., Arychuk, M. D., and Page, J. S.: Temporal and spatial distribution of dimethylsulfide in the subarctic northeast Pacific Ocean: a high-nutrient - low-chlorophyll region, Tellus, 57B, 317-331, 2005.