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Interactive comment on “The impact on atmospheric CO₂ of iron fertilization induced changes in the ocean’s biological pump”

by X. Jin et al.

X. Jin et al.

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Reply to Reviewer 2 (Anand Gnanadesikan)—Part 2: Comments and responses: point by point

Comment: *This paper presents studies of the impact of carbon sequestration induced by "patch" fertilization of the surface ocean. Such projects are currently being proposed by a number of companies, one of which is actually selling voluntary carbon offsets online. Given the price of carbon on the European market, such offsets could easily sell for hundreds of millions of dollars. It is thus extremely important for studies like this to be done that evaluate the impact and efficiency of carbon sequestration. More importantly, the previous study of patch fertilization discussed here by Gnanadesikan*

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et al. (2003) used two very unrealistic models of production, while this paper uses a realistic prognostic model with an explicit iron cycle. Consequently, the results are quite important and deserve to be published. However, I have some important objections to interpretations made in this paper that I feel have to be addressed if the paper is not going to end up being misused by those who have a vested interest in claiming more credit for fertilization than it deserves.

Reply: We very much share with this referee the concern that our conclusions could be misused by a growing commercial community who wants to use iron fertilization as a mitigation option. Therefore, we are emphasizing even more than before that the purpose of this paper is to identify and describe the processes that control the atmospheric uptake efficiency given an increase in biological export production. With this paper, we do not want to get drawn into the discussion whether iron fertilization is a good or a bad mitigation option. In this currently heated debate, our aim is to provide new insights, to show our results (which we believe are very interesting - an opinion shared by all reviewers), and discuss them in order to develop an understanding of the Earth system, so that in the future, we can evaluate all options with the best available knowledge. We believe that a paper in Biogeosciences is not the right place to discuss the political aspects of iron fertilization as a mitigation option, and that should be done through other means, such as the policy forum article of Buesseler et al. (2008).

We also would like to stress that the relevance of the atmospheric uptake efficiency extends well beyond the issue of artificial iron fertilization, as it is helpful to assess the atmospheric impact of any change in export production, such as in response to enhanced nutrient supply from the atmosphere (N or Fe), changes in upwelling and mixing as expected from climate change.

Of course, there is always a possibility that our conclusion will be misused. However, we believe that we have done as much as we can to prevent this: First, we clearly emphasize that the total amount of carbon that can be drawn down from the atmosphere is rather small (last sentence of abstract). Second, we also clearly state and discuss

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two important limitations of our study, i.e. that we don't consider an interactive atmosphere and that we consider the effects over only 10 years. In response to the referee's comment, we have strengthened this latter point in the paper.

Comment: *My major issue with this paper is the interpretation of the results. One can think conceptually about the impact of any perturbation to the carbon cycle in the following terms. Suppose that the perturbation results in increasing the carbon concentration in some locations and decreasing it in others. One can then write*

$$\delta DIC^{ocean} = \delta DIC^{pos} + \delta DIC^{neg} = \frac{\partial DIC}{\partial pCO_2} \delta pCO_2^{pos} V^{pos} + \frac{\partial DIC}{\partial pCO_2} \delta pCO_2^{neg} V^{neg} \quad (1)$$

where the superscripts pos and neg refer to regions where the partial pressure of CO₂ increases or decreases and the V^{pos} and V^{neg} represent the volumes of these regions. The promise of fertilization is that by drawing down nutrients one can create a region of negative anomaly near the surface that will be damped out by the atmosphere, while the positive anomaly at depth will persist over time.

However, there are a number of ways by which this process could be short-circuited. First, the positive and negative anomalies could mix together before equilibration with the atmosphere has a chance to occur. Second, if the nutrients being taken up are "borrowed" from some other point in space and time, positive anomalies will be created at the surface. Third, if the additional production results in denitrification, some carbon will be released when low-nitrate, high carbon water comes to the surface. Fourth, if V^{neg} is large enough, pCO₂ will be very small and the bulk of the carbon uptake will have to come from the ocean.

A key point of this paper seems to be that the fourth mechanism results in a substantial difference between the atmospheric uptake efficiency resulting from enhanced production over the top 20m and in the lower part of the mixed layer. I find it difficult to believe

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this result for the simple reason that the volumes involved seem to me to be far too small. If one were to conserve carbon in the system and include an atmosphere with variable carbon dioxide the equation above would change to

$$R_{C:Air} M_{atm} \cdot dpCO_2^{Atm} + B^{pos} \delta pCO_2^{pos} V^{pos} + B^{neg} \delta pCO_2^{neg} V^{neg} \quad (2)$$

where B is the buffer factor, $R_{C:air}$ is the mass ratio of carbon to air and M_{atm} is the mass of the atmosphere. The point is that if the pCO_2 changes in the negative region come into equilibrium with the atmosphere, the fraction taken from the atmosphere will go as

$$F_{ocean} = B^{neg} \cdot V^{neg} / R_{C:Air} M_{atm} \quad (3)$$

But we know that if we add carbon to the atmosphere, 85% of it will end up in the ocean. So even if the entire ocean buffer capacity is mobilized 15% of a carbon perturbation to either system will end up coming out of the atmosphere. For the tiny volumes here, almost all of the carbon must therefore come out of the atmosphere- if the atmosphere can see the perturbation in time.

Reply: Apparently neither this reviewer nor reviewer 3 (Olivier Aumont) is convinced that we have identified the right process that controls the atmospheric uptake efficiency, i.e. that the depth where the export stimulus occurs is critical in controlling this parameter. Rather, they suggest that the atmospheric uptake efficiency is controlled by downstream effects, in particular changes in the relative volumes of where inorganic carbon is lower than in the control case, and where it is larger.

In particular, this reviewer proposes here an equilibrium scaling argument for why a change in surface water pCO_2 will have minimal effects on atmospheric CO_2 , but the argument as presented is flawed. In fact, as we show below that the correct scaling calculation in fact fully supports our findings.

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The comment "But we know that if we add carbon to the atmosphere, 85% of it will end up in the ocean" is presumably based on some of the very long, quasi-equilibrium findings on fossil-fuel release from the numerical model studies of Archer and others. It holds true only on time-scales equal to or longer than the ocean overturning and mixing time-scales (10^3 - 10^4 years) when the entire ocean volume is equilibrating with the atmosphere. Our focus is on transient responses for much smaller volumes of the surface ocean and for annual to centennial time-scales. "A simple scaling argument can be used to elucidate the thermodynamic limits of air-sea CO₂ uptake and the response of atmospheric CO₂ to a negative surface pCO₂ anomaly. The results of the analysis depend upon the spatial size of the ocean anomaly patch and thus will also depend on time-scale as the patch spreads over a larger volume with time. Consider the case where there is an initial anomaly δP_O^0 in a surface water volume V_O following a biological event. The corresponding initial DIC inventory anomaly in the upper ocean δI_O^0 would be:

$$\delta I_O^0 = \beta V_O \delta P_O^0 \quad (4)$$

where $\beta = \partial \text{DIC} / \partial \text{pCO}_2$ is the buffer factor. Prior to any air-sea exchange, the atmosphere partial pressure and inventory anomalies are zero, $\delta P_A^0 = 0$ and $\delta I_A^0 = 0$. The surface ocean volume is then allowed to equilibrate with a well-mixed atmosphere until the final atmosphere and ocean partial pressure anomalies match:

$$\delta P^f = \delta P_O^f = \delta P_A^f \quad (5)$$

By mass conservation, the ocean and atmosphere inventory anomalies caused by gas exchange must sum to zero:

$$\delta I'_A + \delta I'_O = 0 \quad (6)$$



The inventory changes due to gas exchange for the two reservoirs are given by:

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$$\delta I'_A = \gamma \delta P_A^f \quad (7)$$

$$\delta I'_O = \beta V_O (\delta P_O^f - \delta P_O^0) \quad (8)$$

where γ is a scaling factor relating the change in global atmospheric carbon inventory to a change in atmospheric partial pressure. Combining the last four equations we can solve for δP^f in terms of the initial ocean perturbation:

$$\delta P^f = \delta P_O^0 \cdot \frac{\beta V_O}{(\beta V_O + \gamma)} \quad (9)$$

The fraction of the initial surface water carbon inventory that is replaced by the atmosphere is:

$$F = \delta I'_A / \delta I_O^0 = \frac{\gamma}{(\beta V_O + \gamma)} \quad (10)$$

When the volume of ocean is small relative to the atmospheric carbon inventory, the final atmospheric and ocean pCO₂ perturbations are small relative to the initial oceanic perturbation, and at equilibrium almost all of the carbon initially removed by biology comes from the atmosphere. That is as $V_O \rightarrow 0$, $\delta P^f / \delta P_O^0 \rightarrow 0$ and $F \rightarrow 1$.

Applying some rough numbers, $\gamma \approx 0.2 \cdot 10^{15}$ mol C/uatm CO₂, $\beta \approx 0.6 \cdot 10^{-3}$ mol C/uatm CO₂ / m³, and $V_O \approx 4 \cdot 10^{13}$ m³ for our standard analysis box down to a mixed layer depth of about 50 m. Using those numbers, $\delta P^f / \delta P_O^0 = 1.2 \cdot 10^{-4}$ and $F \rightarrow 0.9999$. On the annual to decadal time-scales relevant to this analysis, the perturbed surface water patch size is small enough that we can treat the atmosphere as

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an infinite CO₂ reservoir that can, given sufficient time, fully replenish the surface water DIC removed by biology as long as there is adequate exposure to the air-sea surface. Lower efficiencies reflect kinetic effects due to finite air-sea exchange, subduction of surface water away from the surface and reemergence of the positive subsurface CO₂ anomalies caused by enhanced subsurface organic matter remineralization. Even for the entire surface Pacific (case X-Large) where $V_O \approx 8 \cdot 10^{15} \text{ m}^3$, the atmosphere still acts as a vast, small changing reservoir, $\delta P^f / \delta P_O^0 = 0.02$ and F => 0.98. Only on time-scales equal to or longer than the ocean overturning and mixing time-scales (10³-10⁴ years) when the entire ocean volume $V_O \approx 1.4 \cdot 10^{18} \text{ m}^3$ ($\delta P^f / \delta P_O^0 = 0.8$ and F => 0.2) equilibrates with the atmosphere does the partitioning of the carbon inventory between the atmosphere and ocean become a serious issue. The result for the whole ocean, while only a scaling argument and not accounting for variations of β with temperature and atmospheric CO₂ and with carbonate sediment adjustments, is broadly consistent with the very-long equilibrium experiments (> 10⁴y) conducted by Archer et al. (1998) and others, who find that about 85% of a carbon perturbation injected into the atmosphere will end up in the ocean (but only on very long time-scales not relevant to our discussion).

In response to this discussion, we have more clearly emphasized the time-scale over which our results apply, i.e. decades, rather than the millennial time-scale of the entire ocean.

Comment: *It is also stated that this effect, caused by production occurring deeper in the water column, is responsible for the low atmospheric uptake efficiency seen in Gnanadesikan et al. (2003). This is incorrect. As can be seen from Figure 14 of that paper, the big impact of simulating iron fertilization a pulse of nutrient depletion was that production was borrowed from subsequent months. The resulting drop in biological productivity over these months meant that no substantial change in preformed nutrients occurred and the uptake efficiency as defined relative to the initial pulse of export was low the runs. That this is the case can be easily verified (and was in the paper) using a*

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simple box model. The definition made in Gnanadesikan et al. (2003) was intentional. To this day there are proponents of fertilization who assert an equivalence between export from the initial bloom and carbon sequestration. A major point of Gnanadesikan et al. (2003) was that such an equivalence could not be made because production could be borrowed from other times and places. In terms of Jin et al., such a naive definition of fertilization would confound the atmospheric uptake efficiency with the iron utilization ratio. In Sarmiento et al. (2007) it is shown that when the iron added to the system is immediately lost, the iron utilization is quite different than when it is retained but the physical-chemical efficiency (defined as it is here) is the same. Further evidence that it is not the depth per se that is involved in producing low efficiencies can be garnered from the fact that the nutrient addition runs reported in the same paper had very high efficiencies- even though the nutrient was added over the same top three boxes (85m).

Reply: We will start with an investigation of the definition of the iron fertilization efficiency used by Gnanadesikan et al. (2003) (referred to as GNAN in the following discussion) and comparing with the definition used in our paper. Then we will give an explanation of the large differences between the two cases in their paper. Finally, we will estimate the corresponding atmospheric uptake efficiency in the two mentioned cases of GNAN, see if their results agree with our conclusions, and analyze what their Figure 14 implies for the atmospheric efficiency.

The physical efficiency used in GNAN is different with the atmospheric uptake efficiency used in our paper and can be written as:

$$e_{phy}(Gnan.) = \frac{\int_t \Delta\Phi_{air-sea}^{CO_2}}{\int_{t_{fert}} \Delta\Phi_{export}^{C_{org}}} \quad (11)$$

Where (t) and (t_{fert}) represent the time intervals, over which the integration is performed. For GNAN, this was usually $t=100$ years, and $t_{fert} = 1$ month. Based on the

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definition used in our paper, we have

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$$\int_t \Delta\Phi_{air-sea}^{CO_2} = e_{uptake}(t) \cdot \int_t \Delta\Phi_{export}^{C_{org}+CaCO_3} \quad (12)$$

$$\int_{t_{fert}} \Delta\Phi_{export}^{C_{org}+CaCO_3} = R_{iron\ util}^{C:Fe}(t_{fert}) \cdot \int_{t_{fert}} \Delta\Phi_{fert}^{Fe} \quad (13)$$

Combining eq. (13) and (12) into eq. (11) and disregarding the export contribution by CaCO₃ we get:

$$e_{phy}(Gnan.) = \frac{e_{uptake}(t) \cdot \int_t \Delta\Phi_{export}^{C_{org}+CaCO_3}}{R_{iron\ util}^{C:Fe}(t_{fert}) \cdot \int_{t_{fert}} \Delta\Phi_{fert}^{Fe}} \quad (14)$$

$$= e_{uptake}(t) \cdot \frac{\int_t \Delta\Phi_{export}^{C_{org}+CaCO_3}}{\int_{t_{fert}} \Delta\Phi_{fert}^{Fe}} \cdot R_{iron\ util}^{C:Fe}(t_{fert}) \quad (15)$$

$$= e_{uptake}(t) \cdot R_{iron\ util}^{C:Fe}(t) \cdot \frac{1}{R_{iron\ util}^{C:Fe}(t_{fert})} \quad (16)$$

$$= R_{iron\ util}^{C:Fe}(t) \cdot \frac{1}{R_{iron\ util}^{C:Fe}(t_{fert})} \quad (17)$$

Therefore the efficiency defined by GNAN is equivalent to the carbon-to-iron fertilization ratio normalized by the iron utilization ratio during the period of the iron fertilization for short term iron fertilization and is equivalent to our atmospheric uptake efficiency for continuous fertilization, i.e. when t_{fert} = t.

Let's consider two iron retention schemes in a simple 1-month fertilization experiment, . In one case the added iron is retained in the system (corresponding in some sense

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to the macronutrient addition case in GNAN and in the other case the iron added is lost from the system (corresponding in some sense to the macronutrient depletion case). As demonstrated by Sarmiento et al. (in preparation) (where the reviewer is a co-author), the atmospheric uptake efficiency, e_{uptake} is similar for these two cases. After the first month, the iron utilization ratio, $R_{iron\ util}^{C:Fe}(t_{fert})$, should also not be very different in these two cases due to high iron concentrations in the patch, assuming iron retention is not important for local NPP in this particular month. Based on eq. (16), the ratio of the efficiency (defined by GNAN) in these two cases is equivalent to the ratio of the iron utilization ratio of them, which is quite different in two cases. Hence, the iron retention can explain the large differences of the efficiency between the macronutrient addition and depletion cases found by GNAN (see Sarmiento et al (in preparation) for details).

The atmospheric uptake efficiency for the two extreme cases of the nutrient addition and depletion can be estimated from Table 3 of GNAN. The third column of the table lists the inverse of the atmospheric uptake efficiency. The ten year efficiency for the nutrient addition case (ONETIME +ADD+EXP) is 37% and the corresponding value for the nutrient depletion case (ONETIME +FULL+EXP) is 25%. We interpret this difference as being due to differences in the vertical distributions of export production. In the nutrient addition case, the nutrient concentrations remain relatively constant over the time, and a higher fraction will come from the near-surface layers. While in the case of the nutrient depletion, the nutrient concentrations will decrease with the time and the closer to the surface, the closer to zero the nutrient concentrations are, resulting in a smaller fraction of export coming from the near-surface ocean.

GNAN identify three separate temporal regimes to show the different impacts of iron fertilization (Figure 14 of their paper). As we have discussed above, their conclusion is true for iron utilization, but for the atmospheric uptake efficiency the story is different. During the first regime (first month), the atmospheric uptake efficiency is very small, the gas exchange flux divided by the biological drawdown of carbon (1.47 MtC/41.9 MtC

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= 3.5%). With the decrease of organic carbon export and increase of air-sea carbon flux in the rebound regime, the atmospheric uptake efficiency should have a substantial increase. And finally the efficiency will arrive at about 25% for ten years.

Comment: *It is important to make this distinction between the operational definition of fertilization and the actual efficiency. Clearly if there is additional export from the surface ocean there will be a lot of uptake from the atmosphere. But the additionality has to be properly defined and more importantly, measured. I think many of those involved in the field have come to understand this, but not everyone has.*

Reply: Is the referee suggesting here that we should alter our definition of the atmospheric uptake efficiency? We would clearly disagree. In particular, we think that both increases and decreases in biological export (i.e. those that occur during the "rebound" period after an iron fertilization event) needs to be considered when thinking about the impact of changes in the biological pump on atmospheric CO₂. Nevertheless, already our submitted manuscript listed the physical efficiencies in the sense of Gnanadesikan et al. (2003). It turns out, that the two definition are highly correlated with each other with an R = 0.6, so that our main conclusion would not be altered had we chosen Gnanadesikan's definition.

Comment: *So why is the efficiency so much lower in Jin et al. when production is increased lower in the water column? Which of the mechanisms outlined above is likely to be important? I would argue that the evidence as presented here is that it is borrowing again. Examining Figure 8 seems to show this quite clearly. The entire additional export from the lower mixed layer has come from the upper mixed layer. In terms of preformed nutrients this would imply that the preformed nutrients haven't changed at all.*

Reply: The second argument that this reviewer brings in is one of "borrowing" in the framework of the conversion from preformed into remineralized nutrients. In our opinion, the concept of the conversion of pre-formed into remineralized nutrients is a useful

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one to help understand how changes in the biological pump can alter atmospheric CO₂. However, the usefulness of this concept hinges on the establishment of a quantitative connection between nutrients and atmospherically derived carbon when this conversion occurs, i.e. when a mol of nutrient is converted a stoichiometric number of moles of carbon are taken away from the euphotic zone and transported below the euphotic zone, creating a flux of carbon from the atmosphere. When viewed in the context of our work, this quantitative connection to the atmosphere is very strong, when the nutrient conversion occurs near the surface, while it is weak when this conversion occurs deep down in the euphotic zone. The extent to which our suggested depth dependence is played out will depend on the depth of the mixed layer. In the high latitudes, the depth dependence is very likely smaller, so that the assumption of a relatively constant quantitative association is relatively good (supporting the work of Gnanadesikan and Marinov on this topic). However, what we show is that in the low to mid-latitudes, this association can vary considerably.

In summary, we don't recognize a contradiction between the concept of preformed versus remineralized nutrients and our key argument. The former one is an interesting diagnostics, but is of limited use for making predictions of the atmospheric uptake efficiency. In contrast, our depth index permits us to make clear and quantitative predictions for the atmospheric uptake efficiency on the basis of a simple metric.

While we are not able to see a contradiction, we do see a problem in the reviewer's argument that borrowing from other times and places is a key factor for a two reasons:

First: Borrowing from other times and places is especially important when one does short-term fertilization. Therefore, if we followed the reviewer's argument, one would expect to see large changes between short and long-term fertilizations. This is clearly not the case as illustrated in our Figure 6, where the atmospheric uptake efficiency remained nearly constant between the one-time, repeat, and standard experiments. We can explain this very well with our depth dependence argument as the depth indices are nearly the same for these three cases.

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Second: In order to support our argument, we undertook nutrient restoring simulations with the same model that Gnanadesikan et al. (2003) used (experiments as described by Jin and Gruber (2003)), in which we also altered the depth dependence of the "fertilization" effect, i.e. in the standard case, we restored the nutrients to zero over the entire euphotic zone, while in the "surface only" case, we restored the nutrients to zero only in the top layer of the model. The atmospheric uptake efficiencies of these two simulations after 10 years followed exactly our predictions, i.e. we found a much higher efficiency in the surface only case relative to the standard case (Figure in this response). For the "Surface only" case, production increased primarily in the top layer for both one month and ten years of continuous fertilization, resulting in high atmospheric uptake efficiencies. In the standard case, production increased primarily in the third layer (or deeper layer) regardless of whether we fertilized over one month or ten years. The corresponding atmospheric uptake efficiencies are only 1/3 of those for the "surface only" case. These results agree well with our conclusion in the paper that the depth distribution is the key factor that impacts the atmospheric uptake efficiency. We have to admit, though, that the relationships presented in our paper could not predict the exact values of the efficiencies, a failure we interpret to be due to the extreme iron fertilization scheme use in the restoring model.

Detailed Comment: *The entire additional export from the lower mixed layer has come from the upper mixed layer. In terms of preformed nutrients this would imply that the preformed nutrients haven't changed at all.*

Reply: Does the reviewer mean "euphotic zone" instead of "mixed layer" here? This is a highly critical distinction, as our argument builds on a clear distinction of the two. We also don't agree with his interpretation of this figure. Before we list the main information from Figure 8b, we would like the reviewer to note that the values in the figure represent the changes in the fluxes. Our findings are as follows:

1. The anomalous DIC transport to the deeper layer, which is really a reduction of the mean upward transport, is balanced by a reduced export of organic carbon from the

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upper layer and a increased input of organic carbon by mixing from deeper layer;

2. Decrease of net community production in the upper layer

3. The anomalous export of organic carbon from the euphotic zone is balanced by the anomalous DIC that is transported and mixed from the deep ocean and laterally by horizontal transport.

Therefore, the anomalous export of organic carbon from the lower euphotic zone does not come from the upper euphotic zone. Instead it comes from the deep ocean and by lateral transport. In terms of preformed nutrients, this would imply that the preformed nutrients are changed (we make here the assumption that a nutrient is considered pre-formed when at depth but still within the euphotic zone). In addition, little of the lateral transport DIC comes from the upper euphotic zone because the global uptake of CO₂ is very small (Table 2 in the paper).

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