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Interactive comment on "Climate engineering by mimicking the natural dust climate control: the Iron Salt Aerosols method" *by* Franz Dietrich Oeste et al.

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We deeply thank the reviewer for his time and for his very helpful and important comments. We have modified the manuscript accordingly and we think that this has largely improved our manuscript in particular for the chapter concerning the effects of iron over the continents (chapter 5), and the concluding chapter 10. Please find below a detailed response to each comments point by point.

Comment 1: The English is not adequate in the paper. Reply 1: The article has been read again carefully and many corrections have been carried out and only one dictionary has been used the "American English". Also one of the reviewers kindly suggested sending a "listing of edits to the European English along with a few typo identifications".

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We hope it would be possible to integrate them.

Comment 2: The organization is poor (most of the conclusions talk about the cost-effectiveness of this solution, instead of pulling ideas together from the paper). Reply 2 : Effectively the original section 8 was both a discussion and conclusion. The cost-effectiveness of the solution proposed is now on a separate discussion chapter 9 (pages 61-64) and the conclusion on chapter 10 (p64-67) now gathers the ideas of the article, summarizing the numerous cooling effects found and the pros and cons of the proposal made, in particular by introduction of table 3.

Comment 3: The paper is full of details, but it gives the impression that the authors don't really understand what they are saying, as there are not always the right papers cited, and the latest ideas included in the paper, which could be a consequence of the great breadth discussed in the paper. Reply 3: As will be seen below on the answer to comment 6, the right articles are now cited and corrections are made, in particular about the work by Mahowald et al, and Boyd and Ellwood, Moore et al. In order to support the latest ideas included in the paper, the recent work on anthropogenic iron emissions is completed (in the conclusion, p68), as well as the chapter 5 (p47-53) about iron over land, forests, wetlands and soil. Thanks to the reviewer comments on this chapter 5, the review has been completed and we believe has been greatly improved. Some criticism of the reviewer seems to point to incomplete or improper concentration of details presented in our manuscript. After the ISA effects on atmospheric CH4 and CO2 concentrations, one of the central aims of our review is the try to demonstrate that the most severe consequence of the ongoing climate warming will be the decline of vertical mixing in ocean basins, as a consequence of halocline- and/or thermocline-induced development of stratified water columns. We ought to give a deeper development to this dangerous consequence. According to this aspect we completed our review by the following details (page 3 in the introduction chapter 1): "The severest consequences of such stratification to oceanic ecosystems is the development of anoxic milieu within stratified ocean basins. A recent example of the development of a halocline stratification is the Black Sea (Eckert et al., 2013). There exists a stable pycnocline which coincides with a chemocline, dividing an oxic surface water layer from an anoxic sulfidic deep layer and the basin bottom contains a black sapropel sediment rich in organic C (Eckert et al., 2013). On page 41 of sub-chapter 4.1: Typical marks of past episodes with stratified water columns in ocean basins are black shale or black limestones as sapropel remnants. Stratified ocean basins during the Phanerozoic epoch occurred as a consequence of elevated CO2 levels in the atmosphere. This caused high sea surface temperatures (Meyers, 2014), and as a global consequence: global increase of evaporation, precipitation and as well production of brines of higher concentrations. It has been pointed out, that the increasing melt water run-off from polar and subpolar ice layers may induce the cover of denser ocean water by a melt water layer (Hansen et al., 2016). The generation of increasing precipitation and surface water run-off accompanied with increasing brine production accompanied by elevated surface water temperatures during hot CO2-high climate episodes had just the same consequences in the past geological epochs as we learn from Meyers (Meyers, 2014). Even the largest mass extinction of biota within the Phanerozoic epoch during the Permian-Triassic transition has been induced by high temperatures as a consequence of elevated CO2-Levels which induced the change of a well-mixed oxic to a stratified euxinic-anoxic ocean (Kaiho et al., 2016). What we have to face now is the extraordinary process developing from the recent situation: the combination of the CO2-dependent temperature rise-generated precipitation increase plus melt water increase. Mankind has to find the appropriate tool to stop and lead back this dangerous stratification process.”

Comment 4: The paper does not convince that the feedbacks described will be large, just that they might exist. There are many other impacts NOT discussed that could offset the impacts suggested in the paper. Reply 4: Concerning possible other impacts not previously discussed that could offset the impacts suggested in the paper, the answer is given on reply 8. It is correct that several of the cooling effects cited and explained in this literature review just mention that they might exist, because there is little or no bibliography available and more research is required to study and model their climate impacts and evaluate their feedbacks. For at list two of them (CO2 removal by ocean fertilization with anthropogenic atmospheric iron and CH4 depletion by °Cl and photocatalytic Fe(II)/Fe(II), or by iron imput in the oceans or in terrestrial landscapes and wetlands) the feedbacks described are expected to be large as there are proposed at the end of the manuscript as possible solutions to fight and stop global warming. Currently, methane (CH4) in the troposphere is destroyed mainly by the hydroxyl radical °OH. From 3 to 4% CH4 (25 Tg/yr) (Allan et al., 2007; Graedel and Keene, 1996) become oxidized by °Cl in the troposphere, and larger regional effects are predicted, up to 5.4 to 11.6% CH4 (up to 75 Tg/yr) in the Cape Verde region (Sommariva and von Glasow, 2012) and ∼10 to >20% of total boundary layer CH4 oxidation in some locations (Hossaini et al., 2016). According to known data (Wittmer et al., 2015), we tried to estimate the methane depleting power of ISA in the troposphere (section 2.2). We found promising results. Even if ISA had only this methane depleting ability, without any others, we ought to go on testing it as a climate controlling tool, and these tests can be done first by computer simulations, climate models, outdoors measurements of existing phenomena and some small scale indoors and outdoors experiments. Recently presented research results of Sherwen et al. (Sherwen et al., 2016a; Sherwen et al., 2016b), about the impact of tropospheric halogens in the atmosphere have shown that some VOC hydrocarbons of the troposphere (ethane, propane and acetone) are depleted from 18 to 9% by °Cl oxidation, the rest by °OH oxidation. ISA sunlight photolysis of Fe(III) can multiply tropospheric °Cl content. This and the research results of Wittmer et al. 2015a demonstrate the CH4 depletion power by the °Cl generating ISA method.

Comment 5: I am only able to review limited portions of the paper: particularly the atmospheric aerosol and impact on land and ocean biogeochemistry, and I found these parts of the paper to be incomplete to the point of wrong. Reply 5: Concerning the impact on ocean biogeochemistry, the answer is formulated in the following reply 6. The answer about the atmospheric aerosol is given below on reply 8, and for the impacts on land biogeochemistry, the answer is given on reply 10.

Comment 6: In the introduction, the paper does a poor job describing the state of our knowledge of iron in the oceans. While I agree that we know less about iron than carbon this sentence is a problem: "meanwhile the iron biogeochemical cycle is only described in the ocean by few scientific publications (Boyd and Ellwood, 2010; Mahowald et al., 2005; Mahowald et al., 2009).:" Please correct these citations: Mahowald et la., 2005 and 2009 focus on the atmospheric iron cycle. For example, work by Ken Johnson, Moore et al., 2013 or Sigman et al., 2010 on the importance of iron in the oceans on ocean biogeochemistry in different time periods would be appropriate papers to cite here. Reply 6: Many thanks for these suggestions. The introduction of the manuscript has been improved concerning the description of the state of the scientific community knowledge of iron in the oceans. Many thanks for the corrections suggested about the atmospheric (and not oceanic) iron cycle. The corrections have been introduced in the manuscript by a modification of the incriminated sentence by the following ones on page 6 of chapter 1 (Introduction): “The biogeochemical cycles of carbon, nitrogen, oxygen, phosphorus, sulfur and water are well described in the literature, but the biogeochemical cycle of the Earth’s iron is often overlooked. An overview of the progress made in the understanding of the iron cycle in the ocean is given by several authors (Breitbarth et al., 2010; Raiswell and Canfield, 2012). The current state of knowledge of iron in the oceans is lower than that of carbon, and still numerous scientific publications deal with this topic (Archer and Johnson, 2000; Boyd and Ellwood, 2010; Johnson et al., 2002a, b; Misumi et al., 2014; Moore and Braucher, 2008; Moore et al., 2013; Tagliabue et al., 2015; Turner and Hunter, 2001), meanwhile the iron biogeochemical cycle in the atmosphere is described by fewer ones (Mahowald et al., 2005; Mahowald et al., 2009; Mahowald et al., 2010), on the contrary to the iron biogeochemical cycle in soil and land, as almost no recent publications details the current knowledge of iron in soils and over the landscape (Anderson, 1982; Lindsay and Schwab, 1982; Mengel and Geurtzen, 1986).”

Also in the introduction we have underlined the recent modeling of the importance of iron from anthropogenic sources on page 7: “Recently the major role of soluble iron emissions from combustion sources became more evident. Today the anthropogenic combustion emissions play a significant role in the atmospheric input of soluble iron to the surface ocean (Sedwick et al., 2007). Combustion processes currently contribute from 20 to 100% of the soluble iron deposition over many ocean regions (Luo et al., 2008). Model results suggest that human activities contribute to about half of the soluble Fe supply to a significant portion of the oceans in the Northern Hemisphere (Ito and Shi, 2015) and that deposition of soluble iron from combustion sources contributes more than 40% of the total soluble iron deposition over significant portions of the open ocean in the Southern Hemisphere (Ito, 2015). Anthropogenic aerosol associated with coal burning would be the major bioavailable iron source in the surface water of the oceanic regions (Lin et al., 2015). The higher than previously estimated Fe emission from coal combustion implies a larger atmospheric anthropogenic input of soluble Fe to the northern Atlantic and northern Pacific Oceans, which is expected to enhance the biological carbon pump in those regions (Wang et al., 2015b).”

The authors thank also the reviewer for the suggested citations. We have unsuccessfully searched for “. . .work by Ken Johnson, Moore et al., 2013 Âż: related to “the importance of iron in the oceans on ocean biogeochemistry in different time periods” and we only found two references of these two authors together, dated from 2002. We did not find work from “Ken Johnson, 2013”, but an article from “Ken Johnson, 2014” was not pertinent. We arrived to the conclusion that the reviewer meant another year and found that Âń Kenneth S Johnson Âż has done numerous interesting work on these topics (but not in 2013) and although not always the first author, we cited some articles which are related to the topics of our manuscript and that he co-authored (but without Moore as coauthor). We also found corresponding work from J. Keith Moore et al, 2013 (without Johnson as coauthor) and these works are now cited in several relevant parts of our manuscript. We thank the reviewer for his helpful suggestions that allowed completing our review. We decided not to refer to Sigman et al., (Sigman et al., 2010), because they reduced their work about the vertical ocean cycle to the carbon burial in the ocean basin sediment phase. But this sediment is underlined by thick alkaline bed rock layers of a total volume of 2.3 x 1018 m3 containing olivine and pyroxene mineral phases and the basic igneous rock types as peridotite, serpentinite, diabas and basalt. This rock base below the sediment layer is known as intensive fractured aquifer. The convective water flow through this huge alkaline ocean crust volume is estimated to about 20 - 540 x 103 km^3/yr (Nielsen et al., 2006). The oceanic crust comprises the largest aquifer system of the Earth with an estimated rock volume of 2,3 x 109 km^3 and a fluid volume of 2 % of the total ocean or $\sim$107 km^3 (Orcutt et al., 2011). The total rock volume of the igneous oceanic crust is 6 to 10 times the total volume of the marine sediments (Ivarsson et al., 2016) The upper part of the igneous crust is characterized by extensive fracturing and microbial within the fractures (Bach and Edwards, 2003; Edwards et al., 2012; Orcutt et al., 2011). From insight into the ocean crust it is known, that it is full of veins and openings filled by carbonates (Coggon et al., 2012; Coggon et al., 2010).

Comment 7: People have rejected the idea of iron fertilization of the oceans for many reasons, and this is not well described in the paper. Are you arguing we should go back and debate this? You are not really discussing the state of knowledge of this debate, or countering it, but rather just ignoring the debate here? Reply 7: The limited knowledge about dissolved or even dispersed iron distributions in the ocean confirms the work of Tagliabue et al. (Tagliabue and Dutkiewicz, 2016): their calculation results about the residence time of iron in the ocean differs up to three orders of magnitude between the different published models. Recently arose the fact that iron from source regions can be transported over distances of several thousand kilometers across the ocean basins (Resing et al., 2015). The iron inputs into the ocean regions by atmospheric dust had been analyzed by several authors (Mahowald et al., 2005; Mahowald et al., 2009; Wang et al., 2015a). Boyd and Elwood (Boyd and Ellwood, 2010) report about the balance between iron supply and removal processes within the ocean. It is right that ocean iron fertilization is very controversial and has many opponents. The recent article from Boyd

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and Bressac (Boyd and Bressac, 2016) opens again the debate and the discussion. Also in the latest IPCC report (IPCC 2013: Carbon and Other Biogeochemical Cycles) the ocean iron fertilization is mentioned as a carbon dioxide removal technology. This debate to decide if iron ocean fertilization is “credible and creditable” is out of the scope of this review article but as this method is compared to the ISA method, we completed the manuscript by the following paragraphs (p65). “The idea of ocean fertilization by iron to enhance the CO2 conversion by phytoplankton assimilation came up within the last two decades. Proposed was the mixing of an iron salt solution by ships into the ocean surface. This idea was debated controversial. Example of this debate is the discussion between KS Johnson et al. and SW Chisholm et al. (Chisholm et al., 2002; Johnson and Karl, 2002). Deeper insight into this debate is given by Boyd and Bressac (Boyd and Bressac, 2016). The iron fertilization procedure tests done so far had been restricted to relatively small ocean regions (Boyd et al., 2007; Johnson et al., 2002a, b). These tests produced iron concentrations orders of magnitude above those produced by natural ISA processing which are in the single decadal order of milligrams of additional dissolved iron input per square meter per year. In this sense the ISA method is quite different from “iron fertilization”. As known from satellite views, phytoplankton blooms induced by natural dust emission events from the Sahara, Gobi, and further dust sources there is no doubt about the fertilizing effect of iron. Meanwhile this kind of natural iron fertilization enhancing the transfer of CO2-Carbon into organic sediment carbon via the oceanic food chain seems to be un-contradicted and accepted (Hansen et al., 2016).”

Comment 8: The authors do not seem to realize that if you add iron to aerosols, they will tend to absorb more incoming radiation, and thus warm the planet: so this is the opposite effect you want. Check Sokolik and Toon, 1999. Reply 8: The global aerosols effects are not trivail (Lohmann and Feichter, 2005; Mahowald et al., 2014) and the direct and indirect effects of absorbing aerosols in the troposphere depend of multiple variables and among them particle size distribution, optical depth, altitude, effects on cloud condensation nuclei, etc. (Conant et al., 2002; Nenes et al., 2002). For

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instance the climate response of black carbon (BC) is highly dependent on the altitude of the aerosol: near the surface BC causes surface warming, whereas near the tropopause and in the stratosphere BC causes surface cooling, despite decreasing planetary albedo (Ban-Weiss et al., 2012; Crutzen, 2006). To take into account this reviewer's remark the following paragraphs are added to the manuscript on page 11: "Differing to any natural dust iron-containing mineral aerosol, the ISA aerosol does not contain any residual mineral components like Fe2O3 minerals known as strong radiation absorbers. Previous studies have shown that iron oxides are strong absorbers at visible wavelengths and that they can play a critical role in climate perturbation caused by dust aerosols (Sokolik and Toon, 1999; Zhang et al., 2015). Because the primary ochre colored aerosol particles emitted by the ISA (method I) have small diameters of <0.05 μm and are made of pure FeOOH they become easily and rapidly dissolved within the plume of acidic flue gas. The ISA FeOOH aerosol becomes emitted with the parallel generated flue gas plumes containing SO2 and NOx as sulfuric and nitric acid generators. ISA stays up for weeks within the troposphere before becoming precipitated on the ocean surface. Because of their small diameter and high surface area, the aerosol particles will immediately react with HCl, generated as reaction product between sea-salt aerosol and the flue gas borne acids. The reaction product is an orange colored FeCl3 aerosol: ISA. During day time the sunlight radiation bleaches ISA into FeCl2 and °Cl; at the night time the re-oxidation of ISA plus HCl absorption generates ISA again. The FeCl2 aerosol particles are colorless at low humidity; pale green during high humidity episodes. The day time bleaching effect reduces the radiation absorption of ISA to much lower levels comparing to oxides like Fe2O3."

Comment 9: None of the section 2.1-2.4 convince me that these effects will be significant. I lost a bit of ability to understand after that, but it seems like many of these feedbacks are actually very long term, and not very helpful in the next 30-300 years (ie. Section 4.3: minimizing ch4 emissions from sediment and bedrock: is ch4 release from bedrock really a problem we have right now, or on geologic time scales?) Reply 9: We agree with the reviewer that CH4 release from methane hydrates sediment, per-

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mafrost and wetlands is a problem we have right now and in the very near future, and that CH4 emissions from bedrock is more a problem on geologic time scales. But to give the whole picture we need to depict the process. In order to be clear about these aspects we add the following paragraphs in the manuscript: “The oxygen-dependent life will have problems within a decreased vertical mixed ocean basin because of its decreasing oxygen content induced by climate warming. The additional input of methane bubbles increases the oxygen deficit death zones. Climate models predict declines in oceanic dissolved oxygen with global warming and the climate warming dependent decline of the oxygen content in many ocean regions has meanwhile become manifest (Stramma et al., 2010). Braking or even reversal of this trend by reducing the oxygen depleting methane emissions at least should help to prevent regions within the ocean basins from methane-induced oxygen deficit. Below the oceanic tropospheric regions with ISA sedimentation even the ocean water column becomes elevated in its iron content before sedimentation of the iron-containing particles at the bottom. Any methane molecule independent if it is existent in the sediment or just above in the water phase or excreted by bubbling into the water column further above, becomes oxidized far away before it arrives at the surface of the water column. By the help of iron containing enzymes even sulfate acts as an oxidant by the help of microbes to oxidize methane to CO2 and water. This prevents the water layers above from oxygen loss. Methane oxidation in wet environments by sulfate is done by sulfate or iron(III)-reducing microbes like archaea or bacteria (Basen et al., 2011). Because these microbes use iron-containing enzymes to do their anaerobic methane oxidation processes they act more efficient in iron-rich than in iron-poor environments (Sivan et al., 2011). The iron containing debris fall of ISA-fed dead phytoplankton and phytoplankton dependent food chain links even feeds the methane depleting sulfate reducer community within or near the sediment surface.”

Comment 10: The whole section 4.4 seems totally off base: it is not thought that the iron inputs from Amazon are important but rather the P inputs, and they only operate on geologic time scales. Reply 10: We deeply thank the reviewer for pointing out to

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us this insufficient description. Now the section 4.4 has been completely reviewed and is the full chapter 5 (pages 47-52). Effectively, for instance Okin (Okin et al., 2004), suggested that soil phosphorus turnover times range from 104 to 107 years. But ISA main components are iron and chloride, thus phosphorus is out of the scope of this review. To briefly summarize the paragraphs that have been added in the manuscript on chapter 5: âĂć iron is involved in many important physiological processes in plants and numerous other living organisms; âĂć although iron is only required in small amounts mainly as a micronutrient, and although iron is the fourth most abundant element of the earth's crust, because of solubility and bioavailability problems many living organisms including plants and humans are deficient in iron; âĂć iron deficiency in human beings concerns 2 billion people and induces many heath problems among which anemia; âĂć iron deficiency induced chlorosis in plants occurs mainly in calcareous and/or alkaline soils (30% of the Earth land surface). This problem can be solved by addition of soluble iron complexes to the soil, or by foliar sprays containing mineral iron (for instance FeSO4) or iron chelates (Fe-EDTA among others). FeSO4 and several organic iron complexes like iron-oxalate are known constituents of atmospheric dust, but unfortunately no published work was found about possible effects on plant chlorosis by foliar deposition of soluble iron from atmospheric dust or from anthropogenic combustion sources; âĂć Fe(III) reduction by micro-organisms in peatlands, wetlands, rice paddies, tropical humid forests and other fresh water surfaces reduces the amount of CH4 release, either by inhibition of methanogens or by increasing CH4 oxidation. This is confirmed by experimental work consisting in artificial addition of several types of iron compounds. Unfortunately no published work was found about possible effects of natural dust or anthropogenic iron deposition on CH4 emissions by humid terrestrial landscapes, although some literature exists about the reduction of CH4 emissions by anthropogenic sulfate deposition.

Comment 11: I would recommend to the authors to focus one paper on each section, not just on describing possible mechanisms, but rather on calculating the impacts of ISA for each mechanism, and making sure the impacts are significant. Do a good job,

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get each idea published, and then you can pull them together later. Reply 11: Finding a possible solution to the climate change challenge is rather complex and needs a multidisciplinary approach and the pieces of the puzzle are not independent but connected to each other by a web of interactions. Several experts (for instance (Hansen et al., 2016)) expressed the urgent warning that mankind has only short time left to address and control climate warming. As a consequence, mankind has to rapidly find possible ways of action, including feedback loops of influence, and cycles of causes. Of course, such tools and agents have to be rapidly evaluated against side-effects to ecosystems, human health, and last but not least their economic burdens. We believe that to improve climate models and integrated assessment models of global climate change it is needed to study inter-linkages between all natural and anthropogenic known or expected biogeochemical and physical climate impacts of the different elements studied and iron is an important one to integrate. The Gaia puzzle cannot be solved by using only a microscopic focus to find out and describe every fitting detail separately localized within the heap of puzzle pieces, all the more they are connected to each other by a web of interactions. Instead, a complete picture is necessary with an immediate presentation of all effects and interactions found out so far as a whole. Allowing an overall vision, with a description of the relevance of details and relevant influences as complete as possible is in our opinion the best way to act. This reviewer suggestion aims to the opposite: first, presentation of all puzzle pieces separately and then, after all the individual pieces have been described, at last publish the whole picture of the interactions of each of the separately described parts. In our eyes this recommendation cannot be the optimum solution to do an emergency job within the shortest time. Mankind has not enough time to act in such an inefficient way. To our knowledge it is the first time that as many iron-depending climate interactions are presented together and even if our review might be completed or needs corrections because some other impacts or effects are not discussed in the manuscript and could offset some of the impacts suggested, we are convinced that it is necessary to present all together as soon as possible the details found out. For optimum efficiency, we need to show to the different geosciences' branches the most complete picture of the ISA method and of the iron effects on all the Earth compartments in order to prove the significance of its numerous impacts in comparison to possible alternatives.

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