**A nature-based negative emissions technology able to remove atmospheric methane and other greenhouse gases**

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**Abstract**

Fulfilling the Paris Climate Agreement requires reducing rapidly the new emissions of greenhouse gases (GHGs) to reach net zero by 2050. As some anthropogenic emissions cannot be zero, to compensate them it will be necessary to remove GHGs from the atmosphere. Among possible methods, the Iron Salt Aerosol (ISA) offers unique potential with a range of cooling effects, including removal of methane and several other GHGs, as well as carbon dioxide. As plans to decarbonize the world economy might also have inadvertent warming effects due to the reduction of iron emissions from fossil fuels burning, ISA can be a remedy to this problem. ISA mimic natural processes that caused cooling during the ice-ages over the past million years. ISA can now be trialed and implemented at a climate-relevant scale. (Add rev1). Desk and laboratory studies indicate low cost, easy deployment and high safety and efficacy, all of which can be validated by field trials.

# Introduction

The Paris climate agreement called to limit global warming under 2°C above pre-industrial levels, is the biggest challenge humanity must meet [[1](#_ENREF_1)]. IPCC Integrated Assessment Models present scenarios that combine reduction of GHG emissions with removal of carbon dioxide (CO2) from the atmosphere [[1](#_ENREF_1)] by Negative Emissions Technologies (NETs).

Numerous NETs have been proposed, the majority of them focusing on carbon dioxide removal [[2](#_ENREF_2), [3](#_ENREF_3)]. But non-CO2 GHGs, especially methane, provide nearly 35% of the climate forcing [[4](#_ENREF_4)] (change ref 4 <https://cdiac.ess-dive.lbl.gov/pns/current_ghg.html>) by well-mixed GHGs. The global warming potential of CH4 ranges from about twenty heigh to eighty four times more than CO2 on 100 year and 20 year timescales [[5](#_ENREF_5)], therefore CH4 removal can significantly reduce its climate impact. The Iron Salt Aerosol (ISA) technology described in this article can oxidize CH4 into CO2, while also causing other cooling effects [[6](#_ENREF_6)] and helping climate restauration.

Anthropogenic sources now add about forty billion tons of carbon dioxide to the atmosphere every year, considered as [[7](#_ENREF_7)]. More than a trillion tons of CO2 equivalent (CO2-eq) would need to be removed to restore the climate [[8](#_ENREF_8)] (Rev 1, modify ref 8 to Allen Myles 2009). To remove billions of tons per year [[9](#_ENREF_9)], other proposed NETs generally require huge new infrastructures. The next section explains how the Iron Salt Aerosol biomimicry method for greenhouse gases removal (GGR) could be a simple, safe and quite cheap way to remove dangerous warming compounds including CH4, tropospheric O3, CO2, black carbon (BC), and some stratospheric ozone layer depleting substances, by replicating main natural processes that cooled the planet in Ice Ages [[10](#_ENREF_10), [11](#_ENREF_11)].

Reducing emissions to zero by mid-century, while maintaining energy use, would require zero-carbon energy sources, with economic and industrial pathways to completely end fossil fuel use. But even ending all CO2 emissions would not be nearly enough to prevent dangerous warming [[12](#_ENREF_12)], due to the committed warming effects of CO2 from previous emissions with feedback amplification already underway, possible tipping points [[13](#_ENREF_13)] and because emissions of other GHGs would continue. (Add Modified Rev 2 Q9). Deployment of iron compounds in the atmosphere may be the most rapid, efficient, effective and safest nature-based method to cut radiative forcing due to GHGs and climate forcers like black carbon [[14](#_ENREF_14)]. (Add explanation Rev 2 Q10). (Complete Introduction Rev 2 Q11 and add plan for following sections).

# Cooling and fertilizing effects of existing iron emissions (change title Rev 2 Q12 and Q13) Iron is an important element for all life forms

(Add transition Rev 2 Q12)

## Biological Need for Iron

Iron is an essential micronutrient for many important physiological processes and bio-functions. Plants need iron, for chlorophyll production, photosynthesis, nitrogen fixation, nitrate reduction, while animals need iron for oxygen transport and many enzyme functions. Human iron deficiency can cause anemia, heart failure, ulceration, skeletal weakness and cognitive disorders [[15](#_ENREF_15), [16](#_ENREF_16)].

## Over the oceans

Over large ocean regions known to be High-Nutrient–Low-Chlorophyll (HNLC), iron deficiency reduces plankton productivity. Phosphorus and nitrogen-rich but iron-poor, HNLC regions cover an estimated sixty million square kilometers, 20% of the ocean surface area [[17](#_ENREF_17)].

In the earth's crust iron is the 4th most abundant element, but its solubility and bioavailability are low. Consequently, large areas of the world’s ocean (equatorial and northern Pacific and most of the Southern Ocean) are iron-deficient, as are calcareous alkaline soils where plants suffer from chlorosis as explained in the next subsection.

Primary productivity in HNLC oceanic regions is limited by the availability of iron [[18](#_ENREF_18)]. Desert and volcanic dust contains iron among other nutrients, and dust deposition in HNLC regions induces algal blooms that can be detected by satellites. These blooms consume large quantities of atmospheric CO2. Increases in iron deposition to these HNLC regions has important implications for the carbon budget, with potential to increase productivity and decrease CO2 in the atmosphere, with a global cooling effect [[19](#_ENREF_19)].

Ocean Iron Fertilization (OIF) by ship-based addition of iron sulfate slurry has been proposed [[20](#_ENREF_20)] as a climate engineering scheme but it has several limitations [[21-23](#_ENREF_21)]. Environmental risks potentially associated with OIF include depletion of deep-water oxygen, increasing production of N2O and CH4, disruption of marine ecosystems and alteration of the food chain, promotion of toxic algae species, and rapid organic carbon re-mineralization that causes CO2 to resurface [[24](#_ENREF_24)]. Therefore some researchers fear that the carbon consumed by phytoplankton blooms may return quickly to the atmosphere [[25](#_ENREF_25)]. Other experiments show that diatom blooms cause deep carbon export and storage [[26](#_ENREF_26)] as explained in section 3.

OIF is different from Iron Salt Aerosol in many respects. Iron Salt Aerosol applies a concentration of iron to the ocean 2-3 orders of magnitude smaller than OIF to enhance marine phytoplankton growth, in combination with other cooling processes that take place in the atmosphere and over the continents. The variety, scope and magnitude of the combined cooling benefits of ISA mean that most if not all criticisms of OIF do not apply to ISA.

Carbon export processes in the deep ocean are complex, time-varying and often decoupled from biological production [[27](#_ENREF_27)], making the efficiency of carbon sequestration by OIF difficult to assess. Alongside the small size and number of OIF field tests, the difficulties of measurement explain why OIF experiments have shown widely varying export efficiency. Calculated molar C:Fe ratios (amount of carbon in biomass created to the amount of iron provided) in natural fertilization vary from about 2,400 to 800,000, with severe under-sampling allowing this wide range [[28](#_ENREF_28)]. Studies of naturally iron-fertilized waters found carbon export efficiencies up to ten times [[29](#_ENREF_29)] greater than short-term blooms induced by experimental ship-based addition of iron sulfate [[30](#_ENREF_30)]. New OIF open field trials have been projected [[31](#_ENREF_31)], as well as new iron dust generation methods [[32](#_ENREF_32)].

. (delete de full paragraph? Rev 2 Q16)

Ice core evidence shows past glaciations over the last million years are correlated with high dust periods and with reduced GHGs [[33](#_ENREF_33), [34](#_ENREF_34)],. (Add sentence Rev2 Q17)

Iron also has other effects: in the oceans, nearly 90% of the CH4 produced within the subsea floor is consumed by anaerobic oxidation by iron oxides and sulfate reduction [[35](#_ENREF_35), [36](#_ENREF_36)]. That means that iron compounds in the oceans help to reduce the emissions of CH4. This is also true in freshwater lake sediments [[37](#_ENREF_37), [38](#_ENREF_38)].

As well as natural dust, iron is also present in the atmosphere due to industrial processes, mainly from coal and other fossil fuel combustion [[39](#_ENREF_39)]. These anthropogenic iron sources are much more soluble and bioavailable than natural ones. (Add 2 references Rev2 Q17, such as 41?). Over many oceanic regions, combustion processes are estimated to provide 20-100% of the soluble bio-available iron[[40](#_ENREF_40)]. That means that anthropogenic Fe emissions from the fossil fuel and steel industries already play a major role in providing bio-available Fe to the surface ocean [[41](#_ENREF_41)] and significantly increase the oceanic primary productivity while also depleting some other powerful GHGs, delivering cooling effects that slow the increase of radiative forcing. If fossil fuel emissions are cut, these existing iron aerosols would need to be replaced to maintain existing oceanic productivity. (Add several references Rev2 Q20 and explain conclusions made)

## Over the continents

It is well known that the Amazon rainforest is fertilized by African dust, due both to iron as much as to phosphorus input [[42](#_ENREF_42), [43](#_ENREF_43)].

For plants grown on the alkaline soils (mainly calcareous) chlorosis induced by iron deficiency is the main nutritional disorder [[44](#_ENREF_44)] that cover more than 25% of the earth’s land surface [[45-47](#_ENREF_45)]. Consequently several methods of chlorosis correction are widely practiced in agriculture, such as iron foliar fertilization and use for soils treatments of synthetic chelates of iron [[48](#_ENREF_48)]. Oeste et al [[6](#_ENREF_6)] found that the deposition of desert dust containing iron as well as the deposition of anthropogenic iron salt aerosols released by the steel industry and by burning coal enables increased CO2 uptake from terrestrial plants including trees and crops, as well as from phytoplankton and other marine plants as explained in previous subsection.

This natural and anthropogenic iron deposition also appears to prevent methane emissions: experimental addition of humic acids and iron on wet sedge tundra landscapes significantly suppressed net methane fluxes [[49](#_ENREF_49)], in Arctic peat ecosystems [[50](#_ENREF_50)] and in rice paddies [[51-54](#_ENREF_51)]. Addition of iron salts either inhibits methane emissions or enhances its oxidation rate [[55](#_ENREF_55)]. The anaerobic oxidation of CH4 is important to limit its release into the atmosphere, with iron compounds playing a key role [[38](#_ENREF_38)].

# Iron present in the troposphere

Atmospheric dust supplied high quantities of Fe to the ocean during glacial periods. This stimulated phytoplankton productivity [[56](#_ENREF_56)] and contributed to drawdown of atmospheric CO2 to levels of less than 200 ppm resulting in glaciations [[18](#_ENREF_18), [19](#_ENREF_19), [57](#_ENREF_57)] and demonstrating efficient C-storage. Anthropogenic pollution emissions now contribute to enhance the natural bioavailability of iron in dust [[39](#_ENREF_39), [58-60](#_ENREF_58)], but the current iron level in the atmosphere is only about one third the estimated level at glacial maxima [[61](#_ENREF_61)]. 800,000 years of climate data from deep Antarctic ice cores reveal that during glacial and inter-glacial periods, the atmospheric concentration of CH4 and CO2 correlated with each other and also with dust levels [[61](#_ENREF_61), [62](#_ENREF_62)]. Natural CO2 atmospheric removal and cooling therefore appear in large part to be caused by iron-rich dust increasing plankton productivity.

Why then did the CH4 atmospheric concentration also decrease? In the troposphere,methane is currently predominantly destroyed by OH• (the hydroxyl radical), but atomic chlorine Cl• oxidizes about 1% (REF Wang X, 2019) (Rev2Q23) of the CH4 (5.3 Tg/yr), or 3-4 % according to older estimates [[63](#_ENREF_63)], possibly produced diurnally by iron salt aerosols as well as by some other natural processes [[64](#_ENREF_64), [65](#_ENREF_65)]. The Cl• creates a CH4 sink that might have increased during glaciations [[66](#_ENREF_66)], together with some other processes that reduced CH4 production [[67](#_ENREF_67)]. (add Rev1). The Cl• is regenerated every day by iron(III)/iron(II) photocatalysis [[68](#_ENREF_68), [69](#_ENREF_69)], according to the following reactions:

FeCl3 + *h*υ 🡪 FeCl2 + Cl• (eq. 1): sunlight generates a free chlorine atom from iron chloride

Cl• + CH4 🡪 HCl + CH3• (eq. 2): the chlorine radical breaks down methane

FeCl2 + NO2 + H2O 🡪 FeCl2OH + HNO2 (eq. 3): an atmospheric oxidant like NO2 oxidizes iron(II) into iron(III)

FeCl2OH + HCl 🡪 FeCl3 + H2O (eq. 4): overnight reactions restore the chlorine atom to iron chloride

The photolysis reaction of iron(III) chloride produces atomic chlorine and iron(II) chloride. The reaction of atomic chlorine by methane oxidation to hydrochloric acid and methyl radical is a first step of the total methane oxidation to CO2. The re-oxidation of iron(II) to iron(III) with an atmospheric oxidant such as NO2 is a complicated chemical process, presented in a simplified way in the equations 3 and 4 above.

Several of the atmospheric processes occurring in the lower troposphere with ISA are described in figure 1:

**

*Figure 1: schematic presentation of the ISA actions in the lower troposphere. Reproduced from Oeste et al. [*[*6*](#_ENREF_6)*] with permission.*

These natural chemical reactions involve iron catalyzing the breakdown of methane by chlorine radical atoms, and show that artificially increasing the amount of iron salt aerosols in the troposphere will mitigate global warming in several ways. The principal effects are by reducing the CH4 concentration in the atmosphere, reducing plant chlorosis by deposition of iron on the continents, and inducing phytoplankton growth that enhances the oceanic food web [[6](#_ENREF_6)] and ocean productivity, after deposition of iron on the oceans. Together these and other ISA processes remove CO2 and CH4 from the atmosphere over both long term and short term.

Ozone (O3) in the stratosphere forms the ozone layer that protects the Earth from dangerous UV radiation. But O3 in the troposphere is a major GHG and air pollutant, harmful to human and ecosystem health. Tropospheric O3 destruction by reactive halogen species such as chlorine, bromine and iodine is well documented [[70](#_ENREF_70)]. During events of tropospheric O3 depletion, over areas of several million km2, the surface ozone is completely destroyed in the lowest km of the atmosphere, producing a measurable cooling effect [[71](#_ENREF_71)] and improved air quality [[72](#_ENREF_72)]. The photochemical activation of chlorine and bromine by iron salt aerosols [[69](#_ENREF_69), [73](#_ENREF_73)] is likely to enhance this process of surface O3 removal, with no negative effect on stratospheric O3.

The IPCC estimated in AR5 that tropospheric O3 has provided 0.4 watts per square meter of the increase in radiative forcing since 1750, nearly as much as methane. The ozone forcing is about 22% of the forcing from anthropogenic CO2 [[5](#_ENREF_5)].

In the tropics, halogen chemistry is responsible for a large fraction (nearly 30%) of tropospheric O3 destruction [[74](#_ENREF_74), [75](#_ENREF_75)]. Globally, up to 3-4% of CH4 tropospheric destruction is due to chlorine [[76](#_ENREF_76)]. In some coastal regions that experience polluted conditions the Cl• sink can account for >20% of boundary layer CH4 oxidation [[77](#_ENREF_77)].

The local albedo is increased by some types of aerosols which reflect sunlight radiation back to space, producing a cooling effect. Marine cloud brightening (MCB), consisting in sea salt aerosols injection over the seas, has been suggested to enhance the cooling potential of low altitude clouds, [[78](#_ENREF_78)]. A further climate outcome of tropospheric iron, possibly the greatest [[79](#_ENREF_79)], is enhancing dimethyl sulfide (DMS) production by plankton causing regional cooling by increased albedo [[80](#_ENREF_80)].

In summary, tropospheric iron aerosols [[39](#_ENREF_39), [58](#_ENREF_58)] enable iron(III)/iron(II) photocatalysis under sunlight, generating chlorine atoms [[68](#_ENREF_68), [69](#_ENREF_69)] which destroy CH4 and surface O3, deposition of soluble iron over iron-deficient oceanic regions enhances marine primary productivity [[81](#_ENREF_81), [82](#_ENREF_82)], and deposition of soluble iron over calcareous or alkaline soils enhances plant primary productivity [[43](#_ENREF_43), [44](#_ENREF_44), [48](#_ENREF_48)]. All this indicates that current natural and anthropogenic emissions of iron salt aerosols already cool the Earth by removing CH4, O3, CO2 and other GHGs and climate forcers from the atmosphere, by enhancing albedo and by increasing oceanic and continental primary productivity [[83](#_ENREF_83), [84](#_ENREF_84)], and by inhibiting CH4 emissions from soils.

# The Iron Salt Aerosol (ISA) method

 Consequently, the observations above indicate that by increasing ISA emissions, several natural GHG removal mechanisms can be safely and efficiently enhanced [[6](#_ENREF_6)]. ISA differs from ship-based OIF methods in that its atmospheric cooling effects are likely to be much more immediate and greater than its ocean fertilization cooling effect, enabling effective implementation with much lower concentrations of iron. However, as a nascent technology, small scale scientific laboratory and field tests are needed to further quantify expected effects and demonstrate safety and efficacy.

Oeste et al. [[6](#_ENREF_6)] found up to 12 cooling effects of iron derivatives in the oceans, in the troposphere and on land. For instance, iron derivatives help reduce the amount of airborne soot emitted by combustion processes [[85](#_ENREF_85)]. Fuel additives containing potential ISA precursors iron picrate complex [[86](#_ENREF_86)] and ferrocene [[87](#_ENREF_87)] are commercially available to enhance combustion efficiency, and reduce fuel consumption and soot emissions. Black Carbon (BC) or soot is not a GHG but causes radiative forcing at a similar level to ozone and methane [[5](#_ENREF_5), [88](#_ENREF_88)], consequently BC depletion or removal is useful. By darkening snow and ice surfaces, BC increases the melting of snow packs, sea ice and glaciers, and by forming heat absorbing atmospheric brown clouds [[89](#_ENREF_89)] it has a warming effect [[90](#_ENREF_90)]. The severe health effects of pollution are well known, with BC, Particulate Matter (PM) [[91](#_ENREF_91)] and surface O3 [[92](#_ENREF_92)] causing several million premature deaths annually worldwide, making reducing their levels highly beneficial for human health [[93](#_ENREF_93)].

Rough estimation of the additional ISA quantities needed to reverse global warming has been calculated [[6](#_ENREF_6)] at the equivalent of current anthropogenic ISA emissions by fossil fuel and steel industries: 100,000 to 200,000 tons per year. Several ways to add the proposed quantity of iron in the lower troposphere are presented in Figure 2, without the associated pollution of the current anthropogenic iron emissions. (add Rev1)



*Figure 2: suggested methods to enhance Iron Salt Aerosols and principal effects acting on climate change. Reproduced from [*[*6*](#_ENREF_6)*] with permission.*

Key features of the ISA nature-based solution include:

* ISA copies the natural global cooling process of the ice ages, where iron-rich dust blowing from continents was a main cooling feedback amplifier;
* today the atmospheric input in ocean surface of soluble ISA from the steel industry and fossil fuels already plays a significant role in ocean productivity;
* iron released in the troposphere, preferably above the boundary layer, will be dispersed not only locally but also regionally, achieving greater dilution by dispersion over a larger surface area, resulting in much better plankton uptake than surface-based OIF;
* iron contained in coal is already released in the hot gas plumes from the stacks of fossil fuel power plants, masking to some level climate impacts induced by the carbon emissions in these flue gases;
* as the iron is most effective in the lower troposphere, a range of methods using surface level chimneys or tethered balloons 200 m above the 1 km average troposphere boundary layer are being considered.
* iron can be released by a balloon in a similar way as planned in the "*SPICE project*" [[94](#_ENREF_94)], sprayed (nebulized) from a solution directly, or after combustion of an oil in which an iron organic complex is dissolved.

As stated by Williamson et al. in 2012 [[95](#_ENREF_95)]: “*Twenty years ago, iron-based ocean fertilization looked like a highly efficient process. Based on the C:Fe ratio in phytoplankton [*[*96*](#_ENREF_96)*], it was calculated in the best case scenario that each atom of added iron might enable plankton growth using more than 100,000 atoms of carbon. Current estimates are that even the natural (re-)supply of dissolved iron to iron-depleted waters is at least an order of magnitude less efficient [*[*29*](#_ENREF_29)*,* [*97*](#_ENREF_97)*], whilst experimental treatments – and natural atmospheric inputs, via dust – seem to be several orders of magnitude less efficient [*[*98-100*](#_ENREF_98)*]*”. Even at these lower levels of efficacy; iron fertilization by ISA could be the single most efficient, rapid, safe and scalable contribution to efforts to slow global warming.

Cost estimates of CDR by ISA are given in Table 1 according to several experimental determinations of the Fe:C ratio in plankton communities before or after OIF.

**Table 1:** **ISA Costs.** Selected literature indicates C:Fe ratios in plankton during iron fertilization experiments, illustrating the possible extremely low costs of Carbon Dioxide Removal (CDR) by ISA employing an iron organic complex as precursor. These figures assume material and delivery costs of US$ 35,000/ton Fe as ferrocene. They are for short term CO2 drawdown, not long-term storage, and do not include the effects of ISA on non-CO2 GHGs which could have total cooling power equal to CO2. The molar ratio of Fe and C is given by Fe/C = 56/12 = 4.65, while the CO2/C ratio is 44/12 = 3.67, giving a Fe/CO2 molar ratio of 56/44 = 1.27.

|  |  |  |  |
| --- | --- | --- | --- |
| Author and {reference}Text from article | moles C:Fe ratio | tons CO2 / 1 ton Fe | cost in US$ / ton CO2 removal by ISA method |
| Morel & Hudson [[101](#_ENREF_101)], Anderson & Morel [[102](#_ENREF_102)], Martin [[19](#_ENREF_19)] | 10,000:1to100,000:1 | 1,706,000to75,871 | $5 / tCO2to $0.5 / tCO2 |
| extended Redfield ratio [[103](#_ENREF_103)]106 Carbon: 16 Nitrogen: 1 Phosphorus: 0.1-0.001 Iron | 1,060:1to106,000:1 | 833to83,280 | $42 / tCO2to $0.42 / tCO2 |
| Ho, et al. [[104](#_ENREF_104)] | 124,000:7.5or16,533:1 | 12,990 | $2.65 / tCO2 |
| Boyd, et al. [[98](#_ENREF_98)] | 26,000:1 | 20,428 | $1.70 / tCO2 |
| Maldonado, Boyd, et al. [[105](#_ENREF_105)]intracellular Iron:C ratios of 6.6 µmol iron / mol C | 151,515:1 | 119,047 | $0.3 / tCO2 |
| Twining, et al. [[106](#_ENREF_106)]Iron:C ratios of 6.0 to 14.1 μmol:mol before iron addition and 40 μmol:mol after iron addition | 166,666:1then25,000:1 | 130,951then19,643 | $0.265 / tCO2then$1.80 / tCO2 |
| Sarthou, et al. [[107](#_ENREF_107)]Fe:C ratio mean value of 4.47 μmol:mol | 233,713:1 | 175,774 | $0.2 / tCO2 |
| Abraham, et al. [[108](#_ENREF_108)]3 μmol Fe per mol C | 333,333:1 | 261,902 | $0.13 / tCO2 |
| Buitenhuis, et al.[[109](#_ENREF_109)]zooplankton and marine bacteria have a Fe:C ratio of 2.10-6 | 500,000:1 | 392,854 | $0.09 / tCO2 |

Even recognizing that the C sequestration is only partial, the cost values obtained in table 1 (as low as $0.10 / tCO2) suggest the ISA method (estimated at ~$1/tCO2) could be the cheapest available Negative Emissions Technology (NET), if field trials demonstrate safety [[2](#_ENREF_2)]. Its dilute dispersion means nature-based solution ISA has high plankton uptake, despite the uncertain amount of carbon that will sink to the bottom of the ocean for long term or even permanent removal from the atmosphere. It should be noted that these cost estimates are partial, as the only effect taken into consideration for this cost-evaluation is the OIF effect of ISA, and its additional effects in the troposphere could equal this cooling effect. In addition, ISA effects should be much more rapid, and the growth of phytoplankton in the upper ocean has the added benefit of increasing ocean biomass and food-web even while it returns some carbon to the air.

Expected ISA cooling effects not taken into consideration for the cost estimations made in table 1 include tropospheric CH4 and surface-ozone removal, CH4 emissions reduction, tropospheric Marine Cloud Brightening, enhanced DMS emissions [[110](#_ENREF_110)] and others described at [[6](#_ENREF_6)]. Also, as will be seen in the next section, the calculations in the table above only take into account the iron content of phyto-planktonic cells, and not the iron recycling, nor the benefits to the oceanic food-web.

# Discussion

One of the principal criticisms made of OIF is that only limited amounts of the organic carbon obtained during phytoplankton blooms may reach and stay stored in the deep ocean, because respiration and bacterial degradation of the dead organic matter will rapidly return much of the CO2 to the atmosphere [[25](#_ENREF_25), [95](#_ENREF_95)]. In the two next subsections we show new evidence that several mechanisms allow prediction of higher levels of mineral and organic carbon storage, in solid and dissolved forms. In the next subsections we address the potential side-effects of reductions of anthropogenic iron emissions and the potential human health risks of ISA emissions.

## The oceanic food web, carbon storage and recycling of iron and other nutrients

The world ocean is more than double the total size of the continents. Its abundant surface area, natural energy and nutrients make the ocean the primary potential location for efficient carbon removal and cooling methods. ISA could prove the single most effective ocean cooling method due to its restoration of the natural levels of iron that prevailed in the ice ages. However, the complexity of the oceanic food-web scatters and dilutes the organic carbon produced by addition of iron, making it difficult to measure the particle flux and carbon export to the deep ocean. Despite evidence of iron-rich dust causing significant reduction in CO2 and CH4 levels with effective carbon storage during the glacial periods [[61](#_ENREF_61), [111](#_ENREF_111)], some scientists fear that a large part of the organic carbon of OIF phytoplankton blooms will decompose back into CO2 and return to the atmosphere [[25](#_ENREF_25)]. Consequently they think that carbon sequestration after OIF might be poor or limited to about 1 GtC yr−1 and might also release CH4 and N2O which are powerful GHGs, offsetting the desired effects of CO2 sequestration[[112](#_ENREF_112)].

However, more than half of all anthropogenic emissions of CO2 are now stored in ocean and land sinks. The 2006-2015 average emissions from fuels and industry are evaluated at 9.3  ±  0.5 Gt C  yr−1 and land emissions (from agriculture, wetlands, etc.) at 1.0 ± 0.5 GtC yr−1. From this total, less than half of the CO2 emissions accumulate in the atmosphere (atmospheric CO2 growth of 4.5 ± 0.1 GtC yr−1) [[113](#_ENREF_113)]. Land and oceanic primary productivity mean about one quarter of emitted CO2 is stored in land (land sink of 3.1 ± 0.9 GtC yr−1) and another quarter in the oceans (ocean sink of 2.6 ± 0.5 GtC yr−1) due also to the gas ↔ liquid CO2 equilibrium between the atmosphere and the ocean. Some computer modeling suggests that to restore pre-industrial levels of atmospheric CO2 concentrations it is necessary to remove double the excess CO2 in the atmosphere, because the oceanic CO2 will outgas to the atmosphere [[114](#_ENREF_114)]. But, most of the oceans’ assimilation of carbon is stored in stable compounds, preventing faster ocean acidification by dissolved CO2 and carbonic acid. In the past, massive amounts of organic carbon burial and storage occurred in marine sediments [[115](#_ENREF_115)], and in oxic sub-seafloor sediment as persistent organic matter [[116](#_ENREF_116)].

Marine carbon storage may prove to be well above levels suggested by critics of OIF [[117](#_ENREF_117)], due to the complexity of the marine biogeochemical cycle [[118](#_ENREF_118), [119](#_ENREF_119)] and the oceanic food web structure, as well as, on a very different scale, the oceanic crustal formation discussed in the next section, which is a major permanentactive alkalinity source and long-term carbon storage locality.

In open ocean trials in silica-rich regions, OIF expands plankton communities dominated by diatoms, allowing efficient deep carbon export and storage [[26](#_ENREF_26)]. In silica-depleted regions of the Southern Ocean (SO), OIF experiments stated that the plankton community was dominated by flagellate, releasing slow-sinking short-lived fecal pellets. But efficient conversion from lithogenic mineral to biologically available iron by copepods allows efficient iron recycling and results in a large spatial spread and longevity of blooms, so a major driver for recycling of Fe in the SO is copepod grazing [[120](#_ENREF_120)].

Krill eat phytoplankton on the ocean surface, but then the krill swarms dive deep, down to 1000 m, where they defecate and their feces pellets become an important part of the carbon bottom sediment [[121](#_ENREF_121)]. The iron content in krill rises with the amount of ingested lithogenic particles [[122](#_ENREF_122)] and this iron remains in the marine food web as krill are eaten. In some laboratory experiments, surprisingly, it is when zooplankton (copepods or rotifers) are added to the cultures that phytoplankton biomass is the highest [[123](#_ENREF_123)] and on average, the presence of copepods increased the iron regeneration rates by 48% [[107](#_ENREF_107)].

Similar mechanisms to the iron recycling processes exist for other important macronutrients (N, P) or micronutrients (Zn, etc.). As all species do not have the same overall chemical composition and metabolic needs as their prey, different proportions of the nutrients ingested by zooplankton will be rejected, modifying the composition of the environment by selective recycling of some elements. For instance, copepods are usually less rich in phosphorus and richer in nitrogen than the phytoplankton they feed on: the P diet excess will be excreted, thereby modifying the N:P ratio in the environment, altering the dynamics of the plankton communities [[123](#_ENREF_123)]. As copepod and rotifer zooplankton groups recycle the nutrients differently, further fertilizations will be more efficient in conditions enriched in N or in P, and after proliferation some species will decline, while others feeding from excreta or from dead bodies of the previous ones will prosper.

Phytoplankton, zooplankton, bacteria, viruses, seaweed, krill, fish, sea-mammals and birds participate in the marine food web, which in turn not only supports human consumption of fish, but also maintains the overall homeostatic stability of the world climate [[124](#_ENREF_124)]. The iron in phytoplankton cells is transferred through grazing into the body organs, tissues and muscle of herbivores, to be later consumed and transferred several more times to higher trophic levels [[125](#_ENREF_125)]. Excess iron intake beyond the metabolic demand of pelagic vertebrates is defecated or excreted at large distances from the initial plankton bloom, by fish, whales [[126](#_ENREF_126), [127](#_ENREF_127)], marine mammals [[128](#_ENREF_128)] and seabirds [[129](#_ENREF_129)]. Some of their waste products spread at distances far away from the initial plankton bloom containing high concentrations of recycled bioavailable iron which can be used for new phytoplankton fertilization. Improving marine biosystems through effective addition of iron could strengthen their resilience and biodiversity through both cooling and nutrient processes, and in turn also support global food security.

As early as 1971, Smayda [[130](#_ENREF_130)] suggested that phytoplankton sinking rates can be accelerated by a range of physical and biological mechanisms in situ (aggregation, packaging of cells in fecal pellets, down-welling and density inversion currents) and this view has since been confirmed [[131](#_ENREF_131)]. Typical in-situ carbon export measurements in the oceanic surface layers that detect solid matter (particles and aggregates) sinking by gravity, cannot easily detect dissolved organic matter downward to the mesopelagic level, 200-1000 m below the surface [[132](#_ENREF_132)].

The role of the ocean as a sink for anthropogenic CO2 [[133](#_ENREF_133)] depends not only on the biological pump [[134](#_ENREF_134)], but also on the microbial pump [[135](#_ENREF_135)]. The biological pump begins with primary producers fixing CO2 in the upper ocean. The carbon (dissolved organic matter or biogenic particles) then sinks to the deep ocean. Some of it is re-mineralized to CO2, and stored in deep waters until it is ventilated at the ocean surface over centennial to millennial timescales [[136](#_ENREF_136)].

Contrary to suggestions of rapid out-gassing, the microbial pump transforms **organic matter into** refractory **dissolved organic C, able to stay thousands of years in seawater. The** global ocean's recalcitrant dissolved organic carbon pool is estimated at more than 600 Gt C [[137](#_ENREF_137), [138](#_ENREF_138)].

## The oceanic crust and the bottom sediment

Recent analysis suggests the carbon sequestration resulting from OIF may be much more than critics have argued, with much less deep-sea carbon returning to the atmosphere than previously estimated. In the depths of the oceans, organic carbon, dissolved organic carbon and carbonates are stored in the sediment by several processes [[139](#_ENREF_139), [140](#_ENREF_140)].

The scale of ocean crust aquifers is immense. Up to 100 billion m3 of ocean water are driven through the porous ocean crust by thermal upwelling every day [[141](#_ENREF_141)]. These waters become chemically reduced in their inorganic carbon and salt content by temperatures above 500°C, alkaline silicates, and sulfate and nitrate reducing conditions. The abundant rock olivine induces carbon precipitation at large scale, generating serpentine, magnetite, hydrogen, heat and a volume expansion [[142](#_ENREF_142)]. The serpentinite reaction can reach depths up to 35 km.

According to Shilobreeva et al. [[143](#_ENREF_143)] samples from ocean bottom drilling have shown that within the young oceanic crust, most of the total carbon content can be organic [[144](#_ENREF_144)], transforming HCO3- and CO2 into stable minerals [[142](#_ENREF_142)].

By these and other processes, organic carbon is fixed in the sediments at the ocean bottom [[145](#_ENREF_145)] and within the underlying ocean crust, sequestered by physical and chemical conditions and processes.

One of the most important cooling effects of the ISA method is that by cooling the surface of the ocean it activates the vertical cycling of ocean currents as well as the increased physical absorption of O2 and CO2 [[6](#_ENREF_6)], with potential for the additional O2 to restore suboxic and even some anoxic oceanic regions. This activated transport also moves the dissolved inorganic carbon from the atmosphere to the carbon sinks in the ocean crust and overlying bottom sediment. It also enhances the transport into the deep carbon sinks the carbon generated by the food chain, and dissolved inorganic carbon originating from continental surface run-off.

According to Liu et al. [[146](#_ENREF_146)]: "*Carbon in rocks and its rate of exchange with the exosphere is the least understood part of the carbon cycle. The amount of carbonate subducted as sediments and ocean crust is poorly known, but essential to mass balance the cycle*". This means current accepted estimations of carbon burial within the oceanic crust may be too low, making the permanent storage resulting from ISA more significant.

## Side effects of reducing anthropogenic iron emissions

Numerous studies suggest that iron dust fertilization in the Southern Ocean (SO) during the last million years caused up to half of each cyclic 80-100 ppm decrease of tropospheric CO2 during glacial cycles [[33](#_ENREF_33)]. During glacial periods, dust deposition in the SO was 3-5 times greater than during the warmer periods (the interglacials) [[34](#_ENREF_34)] mainly because the continents were drier. As well, due to glacial physical weathering the flux of soluble iron was enhanced by ∼15-20 times in the coldest periods compared with interglacials [[147](#_ENREF_147)]. Consequently, a side effect of emissions reductions from combustion processes will be to reduce ocean iron levels even further from their current low level, while an effective approach for cooling must put back more iron than is removed. An important factor in calculating the climate impact of fossil fuel emission reduction is that anthropogenic Fe aerosols is today a major source of bioavailable Fe to the oceans [[148](#_ENREF_148)].

Artificial OIF experiments in HNLC regions provided evidence that phytoplankton blooms are generated by iron addition and can produce efficient C-export and storage in the deep ocean [[26](#_ENREF_26)]. The observation of natural Fe phytoplankton blooms induced by continental dust fertilization [[149](#_ENREF_149)], volcanic ash [[150](#_ENREF_150)], melting icebergs [[151](#_ENREF_151)], krill seabed foraging [[152](#_ENREF_152)] and the upwelling of iron-rich deep waters [[153](#_ENREF_153)], as well as short-term in situ experiments of artificial Fe enrichment, conducted in different HNLC regions of the oceans up to 2009 [[31](#_ENREF_31)] allowed to study the effects of iron on marine productivity and ecosystem structure.

For a significant portion of Northern Hemisphere oceans, about 50% of the soluble iron supply is due to human activities, according to model results [[39](#_ENREF_39)].

In the northern Pacific and northern Atlantic Oceans, the high iron emissions from combustion processes and steel industry suggests larger inputs of soluble Fe of anthropogenic origin, than in the southern hemisphere which is likely to enhance the net primary productivity in those regions [[154](#_ENREF_154), [155](#_ENREF_155)].

Some models estimate the current anthropogenic iron emissions to be about 3 times higher than for 1850, but project by year 2100 a 5-fold decrease [[83](#_ENREF_83)] based on transition away from fossil fuels and clean air standards.

Climate change could reduce desert dust sources of iron, while new regulations aimed to improve health and air quality could reduce the soluble fraction of iron transported to the oceans by reducing acid and mineral emissions from the chimneys of fossil fuel power plants [[58](#_ENREF_58)].

Let us imagine what will happen if all the coal power plants are phased out and if all emissions from steel plants are cleaned out, so that there is no more iron, nitrogen oxides or sulfate emissions. The consequences would be that:

* aerosols with cooling effects will disappear: very good for the health of living organisms but bad for climate: inducing global warming [[156](#_ENREF_156), [157](#_ENREF_157)], with estimated surface heating global mean of 0.5–1.1°C;
* global primary productivity will decrease [[58](#_ENREF_58)] over the continents, which will receive less phosphorus [[158](#_ENREF_158)], iron and nitrogen, although perhaps not in the oceans, because the iron dust accumulated during decades and centuries on the glaciers and poles is now progressively reaching the oceans because of ice melting.
* soils and microorganisms will release more CH4 into the atmosphere that will stay there longer, because the reduced iron amounts in the atmosphere destroy less CH4 and because lower sulfur deposition will also enhance CH4 emissions [[159](#_ENREF_159), [160](#_ENREF_160)].

Consequently, reducing anthropogenic iron aerosol emissions [[58](#_ENREF_58)] or reducing sulfuric acidic pollution [[156](#_ENREF_156)] could provide health benefits through the elimination of the co-emitted pollution, but with serious climate counter-effects [[161](#_ENREF_161)]. Deliberate addition of soluble iron to the atmosphere is a remedy for this unwanted effect that should be tested, and if its safety is proved, it should be deployed in order to counteract or outweigh the expected negative climate effects of the removal of iron involved in decarbonizing the economy.

## Human health impact effects of iron salts

While iron particles can be toxic in some forms [[162](#_ENREF_162), [163](#_ENREF_163)], there are many biomedical applications of iron oxide nanoparticles [[164](#_ENREF_164)] and iron is an essential element for human health, with iron deficiency causing anemia and several disorders [[15](#_ENREF_15), [16](#_ENREF_16)].

1. To minimize health risks, initial ISA release should be over oceans and remote areas, with human inhalation a tiny fraction of existing iron dust levels. Even for workers on ships or platforms that will generate the ISA, the height of release will be sufficient to prevent inhalation, and emissions will be rapidly diluted by wind or ship movement. ISA from ships or platforms should only occur during suitable weather conditions.
2. Natural desert dust emissions contain iron and quartz and during storms can distribute thousands of times more iron than the amount of ISA that is envisaged. These natural iron emissions are localized and occur in short episodes and periods. Conversely, ISA is planned to be released potentially all year long in very dilute form, over numerous well distributed locations, mainly over remote regions of Southern Ocean, andpotentially to remove methane buildup in the Arctic. Potential dust inhalation (containing iron and quartz) is orders of magnitude higher for desert dust than for ISA.
3. We propose that ISA should provide the same mass of iron emissions as current anthropogenic iron emissions by combustion processes and the steel or mining industry [[165](#_ENREF_165)]. But whereas these existing emissions are close to populated areas, we plan ISA emissions largely offshore over regions without inhabitants, so human effects of current iron emissions will remain far higher than those from an ISA program. Additionally, iron emissions are not listed as a significant health problem. A 2017 review of the “*Human health and environmental impacts of coal combustion and post-combustion wastes*” makes no mention of negative impacts of iron [[166](#_ENREF_166)]. Fossil fuel burning and other industrial processes also release toxic compounds and heavy metals such as arsenic, lead, mercury, cadmium, etc. Initial ISA generation would deploy iron that is very low in impurities.
4. In densely populated urban areas, metal levels are already high from cars, trucks, buses, trains and tramways that release by friction or abrasion particles from their brakes, the roads asphalt (often made by mixing with clinker residue from fossil fuel power plants or from incineration plants waste), the rails, the tires, etc.
5. Fuel additives containing iron (ferrocene, iron picrate...) are commercially available, registered and authorized [[167](#_ENREF_167), [168](#_ENREF_168)] and can release iron oxides in urban areas upon combustion. These additives are intended to improve health by reducing pollution from soot and BC emissions [[169](#_ENREF_169)]. Also bunker fuel for ships contains some iron which generates iron aerosols over the ocean [[170](#_ENREF_170)].
6. As the iron salt aerosol will be released over the oceans below or just above the marine boundary layer, it will act as cloud condensing nuclei and with humidity the atmospheric life time will be short. Rained-out ISA will mainly be in soluble form (chlorine salt, oxalate complex [[171](#_ENREF_171)], etc.) with very low risk of inhalation.

We estimate that full deployment of ISA would increase iron oxide particles ‘*available’* for inhalation close to urban areas by less than one part per million, overwhelmed by desert dust and transport and energy sector emissions [[172](#_ENREF_172)]. Against this, the potential cooling impact of a scaled ISA program could deliver major health benefits, reducing the likelihood of further climate tipping points being reached.

# Conclusions

The natural elimination of GHGs other than CO2 in the atmosphere mainly results from reaction with the HO• radical and sunlight photolysis. Evidence that photocatalysis can enhance the speed of destruction of GHGs is well known [[173](#_ENREF_173)]. More and more evidence is accumulating that diverse reactions occur on mineral dust aerosols in the atmosphere [[174-176](#_ENREF_174)]. Iron(III)/iron(II) photocatalytic inducement of halogen atoms in the troposphere has been experimentally verified [[69](#_ENREF_69)].

A biomimicry method has been described in this article: proposing the use of ISA to enhance the amount of tropospheric Cl• in order to destroy CH4, surface tropospheric O3 and soot-BC, and then remove CO2 by diluted OIF; and cool the Earth by several other mechanisms such as producing DMS which will in turn act by MCB effect [[110](#_ENREF_110)].

In view of the risks of climate tipping points such as further destabilization of polar and subpolar glaciers, potential massive release of methane by destabilization of oceanic methane hydrates or melting permafrost [[13](#_ENREF_13), [177](#_ENREF_177)], to our knowledge the ISA nature-based solution and photocatalytic processes [[178](#_ENREF_178), [179](#_ENREF_179)] are the only proposed methods that enhance the speed of atmospheric CH4 removal. ISA might also reduce or inhibit the CH4 emissions, while also mitigating other factors that increase the danger of tipping points.

In the near future, higher air quality standards, aimed to improve air quality and human health, will also reduce aerosol emissions and thus atmospheric deposition of nutrients, among them iron, with strong potential to inadvertently accelerate the decline of oceanic productivity and increase the global warming factor caused by absence of iron aerosols.

Although some oceanic compensation will occur by melting glaciers and icebergs [[180](#_ENREF_180)] as the iron dust deposited in them is released, reduction in anthropogenic emissions from combustion processes will also increase atmospheric GHGs levels through reduction of:

* terrestrial iron fertilization [[58](#_ENREF_58)];
* the tropospheric methane sink provided by the chlorine radical, which is part of a iron(III)/iron(II) photo-catalyzed cycle [[69](#_ENREF_69)];
* iron deposition and sulfate deposition over wetlands and humid landscapes [[181](#_ENREF_181), [182](#_ENREF_182)].

Fortunately, rapid progress in the fight to mitigate global warming can be expected thanks to the development of NETs, which all deserve to be fully evaluated and assessed.

**List of abbreviations:**

AR5: 5th assessment report

DMS: dimethyl sulfide

GHG: greenhouse gas

HNLC: high-nutrient low-chlorophyll

IPCC: intergovernmental panel on climate change

ISA: iron salt aerosol

MCB: marine cloud brightening

NET: negative emissions technology

OIF: ocean iron fertilization

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R K de-Richter declare being member of NGO <https://restoreourclimate.earth/> ; F D Oeste declare holding patent WO/2010/075856 “Method for cooling the troposphere” and member of NGO <https://restoreourclimate.earth/> ; R. Tulip declare being webmaster of <http://www.ironsaltaerosol.com/>.

(Reformat all **references** accordingly with the format indicated in the "Instructions for Authors"

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