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New Directions: Enhancing the natural sulfur cycle to slow global warming[☆]

Full scale ocean iron fertilization of the Southern Ocean (SO) has been proposed previously as a means to help mitigate rising CO₂ in the atmosphere (Martin et al., 1990, *Nature* 345, 156–158). Here we describe a different, more leveraged approach to partially regulate climate using limited iron enhancement to stimulate the natural sulfur cycle, resulting in increased cloud reflectivity that could cool large regions of our planet. Some regions of the Earth's oceans are high in nutrients but low in primary productivity. The largest such region is the SO followed by the equatorial Pacific. Several mesoscale (10² km²) experiments have shown that the limiting nutrient to productivity is iron. Yet, the effectiveness of iron fertilization for sequestering significant amounts of atmospheric CO₂ is still in question. However, marine microorganisms not only consume inorganic carbon but also produce and consume many climate relevant organic gases. The greatest climate effect of iron fertilization may be in enhancing dimethyl sulfide (DMS) production, leading to changes in the optical properties of the atmosphere and cooling of the region.

DMS is believed to play a major role in maintaining Earth's climate in the range necessary for life. This presumption is called the CLAW hypothesis after the authors Charlson, Lovelock, Andreae and Warren (Charlson et al., 1987, *Nature* 326, 655–661). Initially increasing atmospheric CO₂ concentration warms the planet, which induces additional productivity of marine phytoplankton that results in a greater production of DMS.

The CLAW hypothesis further states that greater DMS production would result in additional flux to the atmosphere, more cloud condensation nuclei (CCN) and greater cloud reflectivity by decreasing cloud droplet size. Thus, increased DMS would contribute to the homeostasis of the planet by countering warming from increasing CO₂. A corollary to the CLAW hypothesis is that elevated CO₂ itself increases DMS production which has been observed during a mesocosm scale CO₂ enrichment experiment (Wingenter et al., 2007, *Geophysical Research Letters* 34, L05710). The CLAW hypothesis relies on the assumption that DMS sea-to-air flux leads to new particles and not just the growth of existing particles. If the CLAW hypothesis is correct, the danger is that enormous anthropogenic inputs of CO₂ may overcome Earth's natural ability to self-regulate. Here we present initial calculations and hypotheses intended to spark future, more comprehensive research in amplification of the natural sulfur cycle, aerosol growth and formation, and its possible outcomes.

During the Southern Ocean Iron Enrichment Experiments (SOFeX), we observed seawater concentrations of DMS about five-fold higher inside the iron fertilized patch (7.7 nM; 7.7×10^{-9} moles l⁻¹) versus outside (1.6 nM), 28 days after the initial fertilization (Wingenter et al., 2004, *Proceedings of the National Academy of Science* 101, 8537–8541), while undergoing an approximate three-fold dilution in geochemical signal as reported by Coale et al. (2004, *Science* 304, 408–414). During similar SO iron fertilization projects Turner et al. (2004, *Geophysical Research Letters* 31, L14307), observed comparable enhancements of DMS.

If approximately two percent of the SO were fertilized to about 1 nM of iron three times over the

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course of a week, in well-spaced streaks on the order of 2 km wide, a three-fold dilution in iron similar to that observed during SOFeX would result in an area of about five percent of the SO. The resulting area would be enhanced in DMS concentrations similar to those observed previously (Wingenter et al., 2004), leading to a 20 percent increase in DMS production integrated over the entire SO. If the fertilized strips were orientated roughly North-to-South, orthogonal to the prevailing winds, the enhanced DMS flux may lead to 20 percent more CCN derived from DMS oxidation.

Approximately 200 CCN cm^{-3} arise from DMS oxidation and 200 CCN cm^{-3} from sea-salt in the SO marine boundary layer (Watson and Liss, 1998, *Philosophical Transactions of the Royal Society B* 353, 41–51). Therefore, the total CCN cm^{-3} would increase from 400 to 440 CCN cm^{-3} , assuming the same sulfate to CCN conversion efficiency for the extra DMS air–sea flux. The cloud albedo (reflectivity) over the SO in the summer is about 46 percent (Watson and Liss, 1998). If the number of particles were to increase over the SO by an additional 40 CCN cm^{-3} (from the additional DMS), the albedo would increase by 0.8 percent (employing an analysis similar to that in Watson and Liss, 1998 and in Twomey, 1991, *Atmospheric Environment* 25, 2435–2442). The amount of solar energy reaching the top of the SO atmosphere during the summer is about 400 W m^{-2} . Thus, an additional 3 W m^{-2} would be reflected back to space. Using a global estimate that each W m^{-2} corresponds to 0.75°C (Hansen et al., 2005, *Science* 308, 1431–1435) the initial fertilization of two percent of the SO would result in a 2°C decrease in temperature over the SO region, and perhaps a few tenths over the entire Southern Hemisphere.

It is difficult to predict the full repercussions of a large, 2°C , cooling over the SO without direct observations or at least full scale coupled ocean–atmosphere–ice sheet model simulations. Cooler surface temperature could lead to some slowing of sea-level rise by (i) slowing sea water expansion; and (ii) cooler temperatures may result in diminished break up of ice near the Antarctic coast and inhibit large ice masses, such as the Western Antarctic Ice Sheet (WAIS), from raising sea level rapidly. Melting of the WAIS would raise the sea level by about 6 m.

To fertilize two percent of the SO south of 50° latitude, about 22 kton of FeSO_4 are needed, assuming a 50 m mixed layer depth, which is within the capacity of the smallest class of tanker ships.

However, to accomplish this at the beginning of the growing season over 30 days, about 30 smaller ships would be needed. Changes in albedo and chlorophyll would be monitored and validated by satellite measurements. Depending on the number of satellite and shipboard measurements and analyses made, the yearly cost of such an operation would be on the order of US \$10 million to \$100 million. Initially, a smaller (perhaps 2 percent of $10^6\text{--}10^7 \text{ km}^2$) test area could be enhanced to demonstrate proof of the concept.

An important caveat to this plan is that uncertainties still exist in the understanding of particle conversion of DMS oxidation products. They can either add to existing particles or create new ones depending on many factors, including the atmospheric temperature and the type and amount of oxidant present (e.g. von Glasow and Crutzen, 2004, *Atmospheric Chemistry and Physics* 4, 589–608). Whether new aerosols form or existing ones grow is a critical factor concerning the viability of enhancing of DMS production to cool the climate. A large-scale experiment, as described here, should greatly add to our understanding of aerosol growth and formation in the SO region.

Even limited iron enhancement over several years may result in significant changes in phytoplankton ecosystems, which could have unknown, or damaging, effects to climate or fishing. For example, the SO is known to be a sink of the stratospheric ozone depleting substance methyl bromide. During SOFeX, we observed that net methyl bromide consumption ceased inside the iron fertilized North Patch.

Another possible risk is the buildup of iron below the mixed layer; but because of rapid vertical mixing south of the Antarctic Front this seems unlikely. This mixing could be perturbed if excessive cooling of, perhaps, several degrees resulted in increased sea-ice around Antarctica which would inhibit air–sea gas exchange. Furthermore, decreased buoyancy flux would also suppress upwelling (Watson and Naviera Garabato, 2006, *Tellus* 58B, 73–87). Therefore, iron and turbulence should be monitored.

The estimates presented here are of first order and present the need for fully coupled ocean–atmosphere–cryosphere modeling studies. This should be followed by a pilot experiment over 1–2 years. If limited iron enhancements, on the order of two percent of the entire SO, are carried out, they should be viewed as temporary measures to slow one or more symptoms of climate change until a permanent solution for controlling greenhouse gas emissions can be implemented.

End Note. As opposed to our plan, full scale iron fertilization of the SO for the purpose of sequestering CO₂ seems unfeasible from the perspective of simply overcooling the SO region by several degrees and the likely inhibition of agriculture on the land masses bordering the SO, not to mention other unintended and unknown consequences of massive iron fertilization.

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