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Key Points:

- Modeled soluble iron deposition variability (1980–2015) is characterized for dust, fires, and anthropogenic iron for the first time
- A new anthropogenic iron transient data set is developed, showing lifetime depends on particle emission size and thus air quality acts
- Anthropogenic trends and wildfire variability are important regionally, and daily variability is larger than seasonal or interannual

Supporting Information:

- Supporting Information S1

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Recent (1980 to 2015) Trends and Variability in Daily-to-Interannual Soluble Iron Deposition from Dust, Fire, and Anthropogenic Sources

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Abstract The iron cycle is a key component of the Earth system. Yet how variable the atmospheric flux of soluble (bioaccessible) iron into oceans is, and how this variability is modulated by human activity and a changing climate, is not well known. For the first time, we characterize Satellite Era (1980 to 2015) daily-to-interannual modeled soluble iron emission and deposition variability from both pyrogenic (fires and anthropogenic combustion) and dust sources. Statistically significant emission trends exist: dust iron decreases, fire iron slightly increases, and anthropogenic iron increases. A strong temporal variability in deposition to ocean basins is found, and, for most regions, dust iron dominates the absolute deposition magnitude, fire iron is an important contributor to temporal variability, and anthropogenic iron imposes a significant increasing trend. Quantifying soluble iron daily-to-interannual deposition variability from all major iron sources, not only dust, will advance quantification of changes in marine biogeochemistry in response to the continuing human perturbation to the Earth System.

Plain Language Summary Iron is a limiting micronutrient for marine phytoplankton growth in many ocean basins. A major source of new iron to the open ocean is via atmospheric deposition and until recently was considered to be associated with mineral dust aerosol. However, growing evidence has shown that pyrogenic (fires and anthropogenic combustion) are equally important sources to many basins. Here, for the first time, we quantify the variability (1980 to 2015) in emission and deposition for all three sources across three model versions for robustness. We find that while dust iron dominates the absolute global deposition magnitude, fire iron is an important contributor to daily, seasonal, and interannual variability and that the anthropogenic iron deposition flux to important ocean basins has steadily increased with time. Characterizing more realistic deposition patterns on both short-term (daily to monthly) and long-term (annual to decadal) time scales will improve understanding of the biogeochemical response to the continuing human perturbation to iron emissions and their ocean deposition flux.

1. Introduction

Iron-bearing aerosol absorbs incoming solar radiation (Matsui et al., 2018; Sokolik & Toon, 1999), could be toxic to human health (Ayres et al., 2008; Ghio & Cohen, 2005; Ye et al., 2018), and, crucially, is an essential new source of this micronutrient for marine primary producers (Martin, 1990; Morel & Price, 2003; Tagliabue et al., 2017). Characterization of spatial and temporal patterns in the atmospheric loading and deposition of iron-bearing aerosol is highly uncertain (Myriokefalitakis et al., 2018), partially due to high episodicity in natural emissions originating from multiple sources and regions (Carslaw et al., 2010). Over the Anthropocene, increasing aridity and land use/cover change alters dust and fire emissions (Mahowald et al., 2010; Pechony & Shindell, 2010; Prospero et al., 2002; Tegen et al., 2004; Veira et al., 2016), both important sources of (soluble) iron (Guieu et al., 2005; Ito, 2011; Jickells et al., 2005; Mahowald et al., 2009). In addition, increasing anthropogenic activity has provided a new (soluble) iron source via the combustion of fossil fuels (Ito, 2013; Luo et al., 2008) and industrial processes (Rathod et al., 2020), as well as enhancing land use sources of aerosols (Ward et al., 2014), although the interplay between land use, climate, and fire is

Kok, Longlei Li, Hitoshi Matsui, Natalie M. Mahowald

complex (Andela et al., 2017; Bowman et al., 2009; Kloster et al., 2012). Furthermore, human activity is also a source of those acidic and organic compounds found to increase iron solubility (soluble/total iron) during transport (Ito & Shi, 2016; Johnson & Meskhidze, 2013; Meskhidze et al., 2003; Myriokefalitakis et al., 2015; Solmon et al., 2009). Consequently, major soluble (bioaccessible; Meskhidze et al., 2019) iron deposition sources differ temporally and regionally with anthropogenic activity being the prime determinant of recent changes (Conway et al., 2019; Hamilton et al., 2020; Ito et al., 2019; Krishnamurthy et al., 2009; Luo et al., 2008; Mahowald et al., 2009, 2018; Matsui et al., 2018; Myriokefalitakis et al., 2015, 2020; Sholkovitz et al., 2009).

Ocean biogeochemistry models respond to high-resolution spatial and temporal evolution of not only physical variables (e.g., temperature and light availability) but nutrient deposition also. Primary production and community structure can respond to changes in iron availability on the order of days to weeks in some regions (e.g., within oligotrophic ocean gyres; Guieu et al., 2014), while carbon export likely responds to changes at the interannual scale (Aumont et al., 2008; Doney et al., 2009). Furthermore, as remote marine observations are sparse and on the order of a day, constraining modeled episodicity in natural aerosols with such measurements requires knowledge of the daily atmospheric state (Guieu et al., 2014). Estimating and understanding daily-to-interannual variability in soluble iron deposition from fires and anthropogenic combustion, in addition to dust, to the ocean is thus important.

Here, for the first time, we simulate high-resolution Satellite Era (1980 to 2015) atmospheric emission, processing, and deposition of iron and its soluble fraction within an Earth System Model containing an intermediate complexity iron processing module (Hamilton et al., 2019; Scanza et al., 2018). We compare trends and variability in three major sources of soluble iron (dust, fire, and anthropogenic combustion), using three versions of the model to test for robustness. A new transient (1980 to 2015) monthly anthropogenic iron (defined as sum of smelting iron and fuel combustion of coal, oil, and wood) emission inventory, based on Rathod et al. (2020), is developed and presented here for the first time.

2. Methods

Three 35-year (1980 to 2015) transient simulations were performed using the Community Atmosphere Model (CAM) and Land Model (CLM), within the Community Earth System Model (CESM). Emission data sets are taken from the Coupled Model Intercomparison Project (CMIP) historical climate pathways. Meteorology was nudged to Modern-Era Retrospective analysis for Research and Applications (MERRA) as using reanalysis winds has previously been shown to improve prognostic dust emission variability within CESM compared with free running or ocean forced climate model simulations (Smith et al., 2017). Differences between reanalysis meteorology data sets can alter modeled interannual dust variability (Smith et al., 2017) and thus dust iron deposition. Aerosols were simulated in CAM6 and CAM5 by the modal aerosol module (MAM) and in CAM4 by the bulk aerosol module (BAM). Table S1 in the supporting information details model specifications for CAM6, CAM5, and CAM4. For robustness we report a single multimodel mean deposition flux, seasonality, and related trends, in the main text across these three varying simulations.

Desert dust in all simulations was modeled via the Dust Entrainment And Deposition module (Zender, 2003), updated to include dust mineralogy (Scanza et al., 2015), brittle fragmentation theory on mineral size fractions (Kok, 2011; Scanza et al., 2015), a physical vertical dust flux theory (Hamilton et al., 2019; Kok et al., 2014), and the enhancement of particle asphericity on dust mass extinction efficiency (Kok et al., 2017) and thus dust aerosol optical depth in the visible band (Table S2). Global annual mean dust aerosol optical depth is tuned to be 0.03 (Hamilton et al., 2019; Ridley et al., 2016).

For MAM, the atmospheric soluble iron flux to the oceans was derived using the Mechanism of Intermediate Complexity for Modeling Iron (MIMI; Hamilton et al., 2019). For BAM, we followed Scanza et al. (2018)—on which MIM is based. Both models include atmospheric iron dissolution schemes, via both acid and organic processing, which are suitable for use in an Earth System Model. In all simulations, dust iron emissions are a product of the mineralogy of the originating soil (Claquin et al., 1999; Scanza et al., 2015) and for soluble iron its fractional solubility (Table S3) at emission (Conway et al., 2019; Scanza et al., 2018). In all simulations, fire iron emissions track fire black carbon (BC) emissions using observed Fe:BC ratios from different biomes (Hamilton et al., 2019). For MAM, we extended the 2010 annual mean anthropogenic iron emission

inventory by Rathod et al. (2020) for 1980 to 2010, at 5-year intervals. Fossil fuel iron emissions are based on coal and smelting, wood, and oil (land and shipping) combustion, with a globally uniform soluble fraction of 2%, 10%, and 38%, respectively, at emission. Smelting iron emissions are dominated by steel production, which has remained relatively constant in high-income regions but seen rapid growth in low- and middle-income countries (Figure S1). Pollution control measures are matched by year for fossil fuel combustion but held at 2000 levels for smelting due to a lack of available data for this sector. Emissions for intermediate years were calculated using bilinear interpolation and post 2010 annual anthropogenic iron emissions track CMIP6 BC emission changes, relative to 2010. For each year, a monthly seasonality was imposed by matching iron emission seasonality from oil (land), oil (shipping), coal, and wood combustion to CMIP6 BC emission seasonality from the transportation, international shipping, energy and industrial, and residential sectors, respectively. For BAM, transient anthropogenic iron emissions (fossil fuel combustion only) retain the original inventory by Luo et al. (2008) for 1996 multiplied by a factor of 5 globally to bring in-line with recent observations (Conway et al., 2019; Matsui et al., 2018). The interannual change in monthly anthropogenic iron emissions for BAM then tracks anthropogenic BC ratios, following Equation 1.

$$Fe_{year, month} = Fe_{1996, month} \times \frac{BC_{year, month}}{BC_{1996, month}} \quad (1)$$

3. Results

Natural emissions and deposition are highly episodic (Clark et al., 2015; Hamilton et al., 2019; Mahowald et al., 2009; Patey et al., 2015), and thus, quantifying the daily modeled distribution is of high value for constraining models to daily observations to reduce bias from extreme events, as well as useful to comparing to ocean biogeochemistry observations, since most experiments are on the order of days to weeks to months, and the ocean biogeochemistry response can be different for different time scales (Aumont et al., 2008; Doney et al., 2009; Guieu et al., 2014). Global land cover change since 1980 is minimal (Figure S2), and both dust iron and fire iron are classed as “natural” iron sources, although in reality can be modulated by human activity as well. Annual dust emissions for all versions of CAM (CAM6: 3,439–3,658 Tg a^{−1}; CAM5: 3,753–5,053 Tg a^{−1}; and CAM4: 1,775–2,324 Tg a^{−1}) fall within the full range (735–8,186 Tg a^{−1}) calculated by CMIP5 dust models (Wu et al., 2020). Satellite Era trends and variability in fire and anthropogenic iron emission and deposition are presented and discussed here for the first time.

3.1. Iron Emissions From 1980 to 2015

Globally, iron emissions are dominated by mineral dust sources (Figure S3 and Table S4). However, differences between dust and pyrogenic iron sources attenuate 1–2 orders of magnitude from total to soluble iron emissions, owing to pyrogenic iron having substantially higher fractional solubility than dust iron at emission (Ito et al., 2019; Schroth et al., 2009). Globally, fire and anthropogenic soluble iron emissions are similar in magnitude, but dust soluble iron emissions are between a factor of 3–5 larger (Figure S3b). Modeled fire iron emissions are more soluble at emission than modeled anthropogenic iron emissions due to differences in the amount of the fuel being consumed, iron solubilities for different fuels, and that fire emissions are more fractionated towards fine-sized particles. Observations compiled in Hamilton et al. (2019) suggest a 33%(4%) iron solubility for fine (coarse) fire iron emissions exists, whereas anthropogenic iron is dominated by coal burning and metal smelting processes (Rathod et al., 2020) with an assumed solubility at emission in this study of 2% for all particle sizes. Statistically significant trends (*p* value ≤ 0.013) exist for all sources over the 35-year period: dust iron slightly decreasing, fire iron slightly increasing, and anthropogenic iron increasing (Table S5). Anthropogenic iron emissions have increased faster than fire iron (Table S5), causing anthropogenic activity to exceed fires as the dominant global pyrogenic iron source since 1999 (CAM6) or 2010 (CAM5), depending on fire data set assumptions (CMIP6 and CMIP5, respectively).

Trends in dust are not well known, but there is qualitative data to compare against at decadal time scales (Figure S5). General observed dust aerosol trends originating from three major Northern Hemisphere (NH) source regions (Evan et al., 2016; Kim et al., 2017; Ridley et al., 2016; Shao et al., 2013; Smith et al., 2017) are as follows: North Africa (decrease in late 20th century then plateauing), Middle East (20th century decrease and 21st century increase), and central Asia (decrease over whole period). Both CAM5 and

CAM4 exhibited skill in simulating the general trends for all examined dust source regions (Figure S5), while CAM6, which has a higher spatial resolution and MERRA2 wind fields, captures only the decreasing trend in central Asian dust.

Three Southern Hemisphere (SH) fire iron source regions are also qualitatively examined (Figure S6). As MIMI traces fire iron to fire-BC emissions, trends match those of the inventory used: CMIP5 (Lamarque et al., 2010) or CMIP6 (Van Marle et al., 2017). The absolute magnitude of CMIP5 emissions are ~50% higher than CMIP6 (1980 = 39%, 1990 = 70%, 2000 = 58%, and 2010 = 36%), but variability is significantly lower as CMIP5 emissions were calculated for the first year in each decade only, as compared to CMIP6 which were calculated for each month. Regional trends in CAM6(4) (i.e., CMIP6) are as follows: South Africa (relatively steady), South America (20th century increase post-1987 and moderate 21st century decrease), and Australia (generally steady with a notably strong early 21st century peak and possible positive post-2010 trend evolving). The fire Satellite Era began in 1997, and uncertainty over the first half of the study period is thus higher than the latter half, although recent emission estimates remain poorly constrained (Pan et al., 2020; Reddington et al., 2016). Confidence in South American twentieth century emission patterns for CAM6(4) is higher than other examined regions as pre-1997 Brazilian fire estimates also incorporated visibility observations (Van Marle et al., 2017).

Like the natural sources, discussed above, we consider three NH anthropogenic iron source regions to examine qualitative emission change between western countries (North America and Europe) and Asian countries (Figure S7). Anthropogenic iron emission trends have broadly followed observed and modeled $PM_{2.5}$ (particulate matter <2.5 μm in diameter) changes (Leibensperger et al., 2012; Tørseth et al., 2012; Turnock et al., 2020). Air quality improvements, through technological advances aimed at reducing $PM_{2.5}$, are counterweighed by population growth and energy demands; in China and India the latter has outweighed the former in recent decades (Daskalakis et al., 2016) and thus when combined with exponential steel production growth (Figure S1) Asian iron emissions rise, while North American and European emissions fall.

3.2. Atmospheric Iron Lifetime Changes and Fractional Solubilities Over Time

Abatement methods for larger particles are more effective and cheaper than for smaller particles, for example, cyclone filters versus bag filters (e.g., Klimont et al., 2002), hence favoring their application. The contribution of coal burning to soluble iron emissions has thus risen slower (1980 to 2010) for coarse particles (8% increase; from 66% to 74%) compared to fine particles (18% increase; from 44% to 62%). Figure 1a shows that while at the beginning of the studied time period, coarse-sized particles were emitted in higher quantities than fine sized, after 2010 they were similar, a transition which occurred some 15 years earlier for soluble iron (Figure 1b). The shifting of the emission particle size distribution toward finer-sized particles increases anthropogenic iron lifetime; a response we propose is related to a decrease in dry deposition loss for this iron source (Figure S8), which favors deposition of larger heavier particles. A secondary impact is an increase in the critical supersaturation required to activate particles to cloud condensation nuclei with reducing particle size, which, all other processes remaining constant, could reduce wet deposition losses also. Extending CAM6 anthropogenic iron atmospheric lifetimes by ~40% (from 1980 to 2015) increases the long-range transport potential and thus the amount of atmospheric iron processing to a soluble form, which can occur. Lifetimes differ between total and soluble iron burdens due to global heterogeneity in fuel consumption (coal, wood, or oil) patterns, and thus differing fuel solubilities at emission create differing regional aerosol burdens undergoing different rates of loss. MIMI accounts for the specific surface area due to composition (mineralogy) but not changes in surface-to-volume ratios with changing particle size distributions during transport; therefore, increasing the relative available reaction surface area could increase dissolution rates further than shown here. Figure 1 thus represents a lower bound of the impact of air pollution controls on anthropogenic iron lifetime changes and dissolution potential.

Acid processing of atmospheric iron aerosol in MIMI is simulated using an intermediate complexity solubilization mechanism, which depends on the relative absolute contribution of the two main simulated species governing aerosol acidity: sulfate and calcite (Hamilton et al., 2019; Scanza et al., 2018). Acid processing of iron is thus related to pollution (Li et al., 2017; Meskhidze et al., 2005; Solmon et al., 2009); however, globally the calcite (sulfate) relative contribution has remained larger than sulfate (calcite) in coarse (fine) mode aerosol over the study period, and thus, the global mean atmospheric iron source fraction from acid

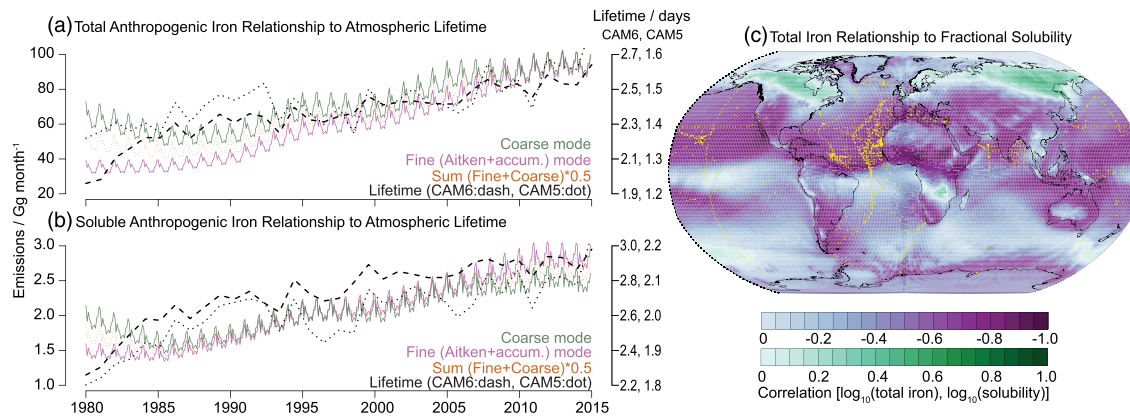


Figure 1. The relationship of anthropogenic iron to lifetime (a and b) and total iron to its fractional (%) solubility (c) over time (1980 to 2015). The annual mean lifetime of total (a) and soluble (b) anthropogenic iron aerosol is partially controlled by particle size at emission. Monthly mean coarse mode ($>1\ \mu\text{m}$; green) and fine mode ($<1\ \mu\text{m}$; purple) emissions shown with halved (to match scale) sum (orange). Annual mean aerosol lifetime shown for CAM6 (black dashed line) and CAM5 (black dotted line). Most iron observations (yellow dots; Mahowald et al., 2009; Myriokefalitakis et al., 2018) have historically been taken in regions of high negative correlation (c). Total iron in (c) is the sum of all sources: dust, fires, and anthropogenic.

processing has remained relatively unchanged from 1980 to 2015 in these simulations. The result being that despite the simulated global mean sulfate burden increasing by 14% over the study period $\left(\frac{2010 - 2014}{1980 - 1984}\right)$ global annual mean coarse (fine) mode pH has remained relatively fixed between 6.4 and 6.6 (1.1 and 1.2). Sholkovitz et al. (2012) used observations to show that the relationship of total iron to its fractional solubility can be described spatially at the global scale by an inverse hyperbolic relationship. Mahowald et al. (2018) used a box model to show that this could be due to either anthropogenic iron sources being finer and more soluble or, focusing on the spatial variability, that atmospheric processing causes longer-lived particles to have higher solubilities farther from their source. Here we examine the temporal relationship at the same location in the model, and we find that the negative correlation between total iron and its fractional solubility (in log-space) also holds across time in most regions, especially where observations have historically been made (Figure 1c). This implies that both spatial and temporal anticorrelations between high iron amounts and higher solubility occurs over those regions which are usually sampled. However, lower correlations are found outside main observational regions, particularly within SH ocean regions, and high positive correlations are found in boreal forest regions.

Simulated CAM6 (CAM5) daily median SH ($>30^\circ\text{S}$) surface level iron solubility per source broadly agree with observations (Table S6) and are as follows: dust iron: 1.4% (1.8%), fire iron: 18.8% (19.6%), and anthropogenic iron 11.2% (13.4%). However, modeled dust iron solubility is highly variable, reaching a single day maximum of 65.0% (44.1%). Long-range dust transport across the Southern Ocean is common (e.g., McConnell et al., 2007), and initial dust iron emission solubility is very low ($<1\%$; Table S3); therefore, a high degree of dust iron processing can be undertaken in the model, consistent with observations in the Southern Ocean region (Heimbürger et al., 2013). Consequently, it is important not to assume Southern Ocean high iron solubilities are solely the province of pyrogenic iron sources (Baker & Croot, 2010; Mahowald et al., 2018).

3.3. Spatial and Temporal Soluble Iron Deposition Variability

We focus the discussion on regional soluble iron deposition variability owing to its impact on marine biogeochemical cycles (Tagliabue et al., 2017). Overall, we find that wet deposition dominates modeled iron deposition fluxes to the ocean (Figure S8), consistent with iron observations (Baker et al., 2010; Chance et al., 2015; Gao et al., 2013). Daily mean cumulative 35-year variability (standard deviation/mean) in deposition is a factor of 3 higher than monthly mean variability (Figure S9). Daily model output frequency is thus required to capture extreme deposition events (Mahowald et al., 2009), improving model comparisons with, often daily, remote marine observation comparisons (Hamilton et al., 2019). Knowledge of daily deposition rates could also support quantification of shorter-term (daily-to-weekly) responses in primary productivity in those

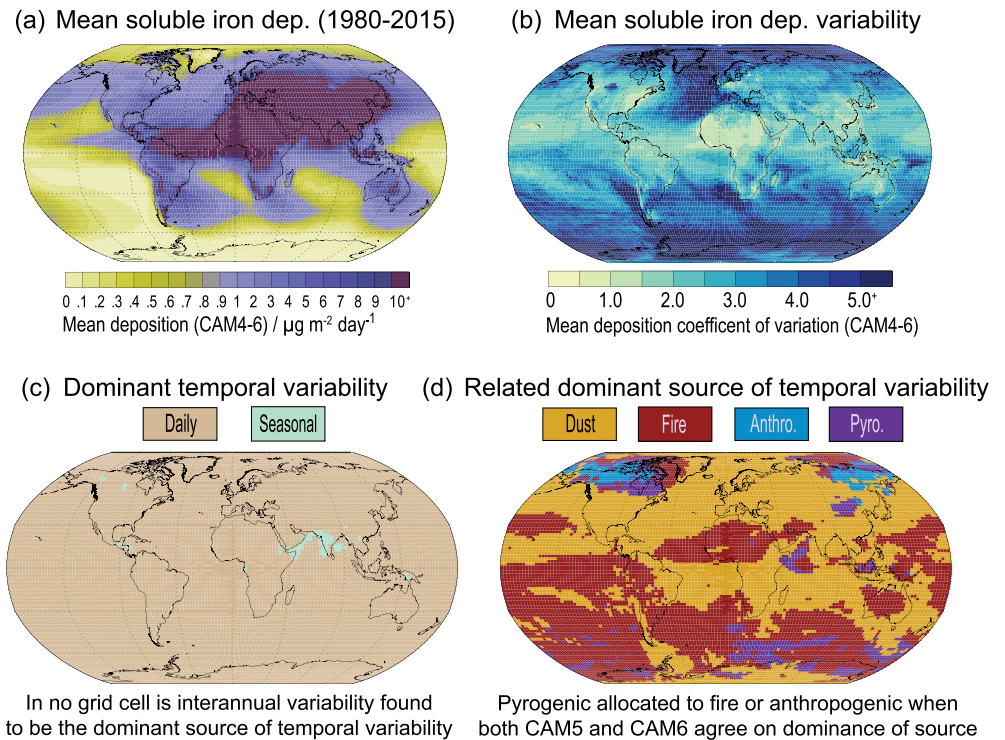


Figure 2. Patterns in soluble iron deposition: 1980 to 2015. Soluble iron deposition climatology (a) and variance (b) (Figure S10: variance for each model and source). Dominant temporal variability (coefficient of variability = standard deviation/mean) (c) and associated iron source (d) (Figure S11: variance for each model).

regions potentially sensitive to episodic nutrient fluxes from natural iron sources (e.g., oligotrophic ocean gyres; Guieu et al., 2014).

As shown previously for dust (Smith et al., 2017), a high soluble iron deposition flux, related to dust iron sources, is associated with low temporal variability (Figure 2a vs. Figure 2b). Analysis that separates interannual variability, the seasonal cycle, and daily variability shows that almost everywhere, daily deposition variability is largest (Figure 2c). Variability is maximal over middle- to high-latitude ocean basins and for the Southern Ocean associated with natural, predominantly fire, iron sources (Figures 2b and 2d). Observations within modeled regions of high SH deposition variability similarly span a large range of iron solubilities (Table S6), and observed high solubilities are proposed to be associated with pyrogenic iron sources (Ito et al., 2019). However, in situ remote marine observations (Figure 1c) are sparse in regions of high variability, normally representing a single-day snapshot of the atmospheric state; therefore, further observations (Meskhidze et al., 2019) are required to isolate possible regional and seasonal iron source covariances with modeling predictions.

Next, we consider the seasonal-to-decadal variability in soluble iron deposition to four ocean regions (Figures 3a–3d) defined by source apportionment, which relates deposition to 10 distinct iron emission regions (Figure S4).

Region 1 is dominated by a strong North African dust iron source (Figure 3a). Summer-to-winter months express a decreasing 35-year trend in dust iron deposition, consistent with observations of decadal mid-Atlantic dust aerosol optical depth and Barbados surface concentrations (Ridley et al., 2014). North African dust iron source reductions are related to reduced surface wind speeds within source regions (Evan et al., 2016), vegetation coverage changes (Kim et al., 2017), and precipitation decreases within the Sahel (Taylor et al., 2017). Seasonality in fire iron follows dust iron with two peaks: in winter associated with a northern Africa fire iron source and in summer associated with a boreal forest fire iron source (Giglio et al., 2010). North African dust flux variability increases away from the major outflow transport plume, suggesting that while dust iron under the plume dominates the total magnitude of the iron flux, it is fire iron

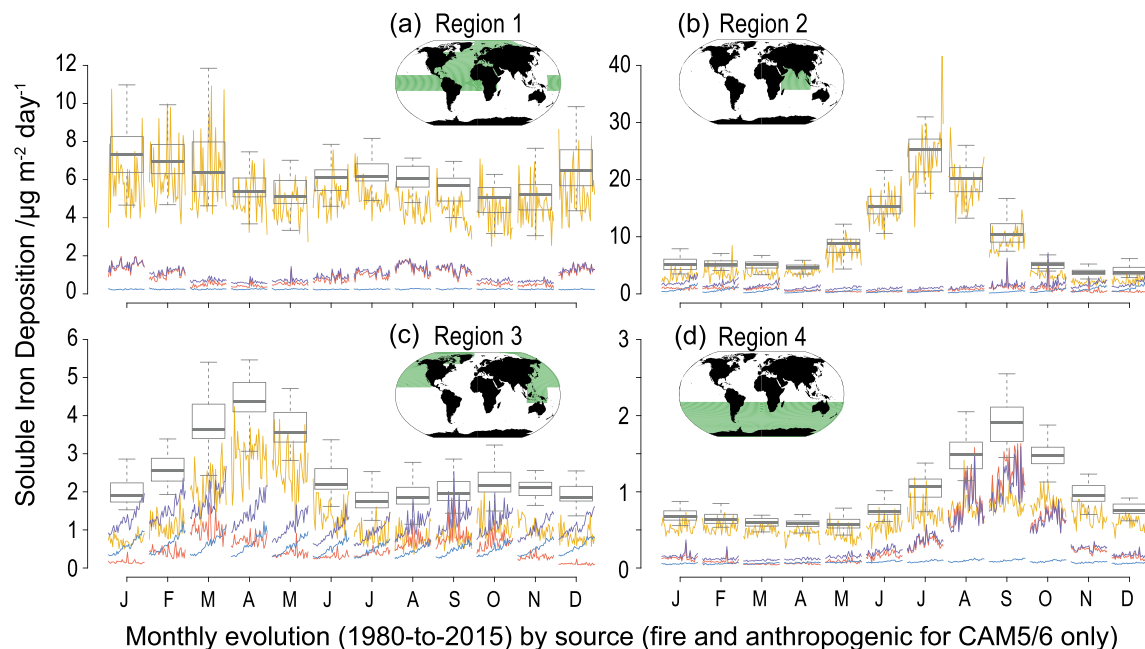


Figure 3. Monthly evolution (1980 to 2015) in the ocean flux to four distinct basins (a–d). Sources: dust iron (gold; mean CAM4:5:6), fire iron (red; mean CAM5:6), anthropogenic iron (blue; mean CAM5:6), pyrogenic iron (purple; mean CAM4:5:6), and all sources (box and whisker). Marine regions delineated by source apportionment, which has been shown to define ocean regions more clearly for iron biogeochemistry studies than a more traditional basin classification, based on physiogeographical location, would (Hamilton et al., 2019). Each data point in the plot for each month represents the value for each year across the 35 years.

that dominates the variability (Figures 2b and 2d). Average anthropogenic iron ocean inputs across the whole region are minimal, but in the northern Atlantic away from dust outflows will be comparatively higher (Conway et al., 2019). Due to the significant North African dust iron source, ocean biogeochemistry modeling suggests that much of this region is not iron limited for most phytoplankton groups (Moore et al., 2013). One exception being the iron-limited equatorial Pacific; however, increasing iron deposition along this nutrient upwelling zone may have minimal impacts on basin-scale net primary productivity because increased nutrient uptake here diminishes that nutrient availability downstream, and thus offsetting patterns of productivity are predicted within the Pacific (Hamilton et al., 2020; Tagliabue et al., 2008).

Region 2 is controlled by the strong East African and Middle East dust iron source (Figure 3b), linked to the summer monsoon (Guieu et al., 2019; Wiggert et al., 2005). Modeled seasonality matches long-term (26 years) aerosol index observations (Banerjee & Kumar, 2016) with maximal dust deposition occurring in July. This highly productive tropical marine region (Roxy et al., 2016; Wiggert et al., 2005) is dependent upon nutrient supply from both upwelling and atmospheric deposition, with dust iron suggested to support at least half the Arabian Sea's primary production (Guieu et al., 2019). Recent rapid ocean warming is enhancing stratification of the water column, diminishing nutrient upwelling, and thus has significantly decreased long-term productivity in the region (Roxy et al., 2016). Opposing the nutrient limitation due to stratification, the dust iron supply is shown here to be steadily increasing in both magnitude and interannual variability from 21.9 ± 3.2 (1980–1984) to $31.0 \pm 8.4 \mu\text{g m}^{-2} \text{ day}^{-1}$ (2010–2014). Interannual variability in dust deposition is linked to the El Niño–Southern Oscillation (Banerjee & Kumar, 2016), and rising air temperatures and aridity tend to increase dust emissions (Kok et al., 2018; Namdari et al., 2018). How far increasing dust nutrient supply could offset stratification nutrient depletion is an important future question for predicting productivity here.

Region 3 is dominated by Asian iron sources, which shows the strongest seasonality of all regions (Figure 3c). Strong spring-to-early summer deposition of dust iron, peaking in April, depends on wind strength and precipitation amount within the originating desert source regions. Westerly airflows mix the

entrained dust iron with pyrogenic iron and pollution, enhancing solubility, as it travels eastward through China and out over the North Pacific. A smaller autumnal peak is also simulated when pyrogenic iron sources have an increased impact on deposition. Increasing Asian anthropogenic activity (Figure S7), especially within China (e.g., exponentially increased in steel production: Figure S1) drives the clear positive trend in anthropogenic iron deposition and is predicted to continue through end of century (Hamilton et al., 2020; Myriokefalitakis et al., 2020). The response of marine productivity in the North Pacific to an increasing anthropogenic iron source is uncertain; two studies suggest increases (Ito et al., 2020; Myriokefalitakis et al., 2020), while a third study shows a small decrease but increasing rates of nitrogen fixation (Hamilton et al., 2020). Air quality control measures along with increasing economic activity in Asia likely increase long-range aerosol transport and iron solubilization (Figure 1), due to an increasing relative fraction of finer aerosol particles, relieving iron limitation within the remote North Pacific Subtropical Gyre. In the future such increases in soluble iron fluxes could push productivity toward longer periods of phosphorus limitation (Letelier et al., 2019).

Region 4 is characterized by SH iron sources (Figure 3d). South America, Southern Africa, and Australia all supply both lithogenic and pyrogenic iron (Myriokefalitakis et al., 2018; Perron et al., 2020). Seasonality is similar for both dust and fire iron deposition with a September peak, possibly as both are anticorrelated to precipitation (e.g., Archibald et al., 2013; Gassó & Torres, 2019), but the seasonal amplitude of fire iron deposition is larger. Fire iron exhibits a rising twentieth century trend subsequently falling to 2015, likely driven by changing rates of deforestation activity in Brazil (Reddington et al., 2015). While the recent (2019) Australian megafires are beyond the investigated time frame, they should have large impacts on future deposition to this region if persisting. The magnitude of soluble iron deposition within the Southern Ocean (50–70°S) is lower than Region 4 as a whole, but the seasonality in iron sources is well matched with the amplitude of fire iron deposition only slightly attenuating in relation to dust iron (Figure S12). This figure supports the growing evidence (Barkley et al., 2019; Hamilton et al., 2019, 2020) that wildfires are likely to be an important contributor of atmospheric soluble iron fluxes to the iron-limited Southern Ocean, presently and under future climate change.

4. Conclusions

Modeled long-term soluble iron deposition generally follows observed emission trends for all major emission sources investigated: dust, fires, and anthropogenic activity. Here we explore these trends using three different versions of the same model, driven by MERRA reanalysis. Dust iron has dominated NH (soluble) iron deposition over the past 35 years. Increased Asian anthropogenic activity and pollution controls fractionate anthropogenic iron emissions toward smaller particle sizes by removing coarse-sized ($>1\ \mu\text{m}$) particles more efficiently, increasing the overall lifetime of anthropogenic iron (and particle surface-to-volume ratios) and hence dissolution potential with time; therefore, we find pyrogenic iron has overtaken lithogenic iron within the early part of this century as the dominant North Pacific soluble iron source outside spring. Over time, the observed inverse hyperbolic relationship describing the relationship of total iron to its fractional solubility (Sholkovitz et al., 2012) is strongest where observations have traditionally been taken in the NH and weakest in remote SH marine regions. Characterizing Southern Ocean deposition is important, owing to carbon export here being an effective loss pathway for atmospheric CO_2 (Kohfeld & Ridgwell, 2009; Martin, 1990), and both dust and fires are important soluble iron source to this region but highly susceptible to anthropogenic activity and a changing climate. That equivalent high variability in modeling and limited observations exists where atmospheric iron effectively modulates the global marine carbon cycle hinders estimating the impact of human activity on marine biogeochemistry and changes to carbon export efficiency, and in this context, more observations are required. Due to logistic expense, however, climate and biogeochemical studies will likely entail significant modeling for the foreseeable future, and thus, quantifying iron emission and deposition variability, at high resolution, from pyrogenic and dust iron sources, as presented here, is essential.

Data Availability Statement

Data is available from the Cornell eCommons repository (<https://doi.org/10.7298/xqqj-qk90>).

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