

The ozone depletion theory of global warming

Peter L. Ward¹

Mean global surface temperatures have remained essentially constant since 1998 while carbon-dioxide concentrations continue to increase. Record high temperatures and drought were common in North America during 2012-2013, while record rains flooded England. Here we show that ozone depletion caused by anthropogenic chlorofluorocarbons and small, effusive volcanic eruptions explains these and other climate anomalies clearly. The highest-energy ultraviolet-B radiation from the sun is normally absorbed by the ozone layer in the lower stratosphere. When ozone is depleted, as observed since 1970, more ultraviolet-B energy is measured reaching Earth. Greenhouse-gas theory underestimates the thermal effects of ozone depletion because it assumes electromagnetic radiation propagates as waves in space. Radiation transfers thermal energy as frequency, not wavelength. The thermal energy in ultraviolet-B radiation is 48 times the thermal energy in infrared radiation absorbed by greenhouse gases. There simply is not enough thermal energy absorbed by greenhouse gases to cause observed global warming.

Mean global surface temperatures changed little from 1945 to 1970, rose sharply until 1998, and have remained nearly constant since (red bars, Fig. 1)¹. Meanwhile atmospheric concentrations of carbon dioxide increased monotonically and are now increasing at the highest rates ever observed². This divergence in trends suggests that increasing concentrations of greenhouse gas may not be the primary cause of global warming.

By 1965, production of chlorofluorocarbon gases (CFCs), used for refrigerants, propellants, and solvents, began increasing rapidly, leading to an increase in total tropospheric anthropogenic chlorine (green line)³. In 1974, scientists discovered that CFCs, when exposed to ultraviolet sunlight in very cold environments in the lower stratosphere, release chlorine atoms that destroy ozone⁴. It takes CFCs three to five years to be moved up into the lower stratosphere⁵. Ozone concentrations in the lower stratosphere began decreasing by 1970 (black line, y-axis reversed)^{6,7}. Discovery of the ozone hole over Antarctica in 1984 added urgency to negotiate the Montreal Protocol on Substances that Deplete the Ozone Layer by 1989. Under this protocol, emissions of CFCs began to decline by 1993, and increases in ozone depletion stopped after 1996. Both the onset of rapid warming by 1970 and the cessation since 1998 can be explained by ozone depletion caused by anthropogenic CFCs (Fig. 2b). CFCs are very stable, persisting in the atmosphere for as long as a century³. Therefore ozone remains depleted, explaining the continuing increase in ocean heat content⁸.

Mean global surface temperatures result from a delicate balance between net thermal energy lost from Earth's surface via radiation and convection versus net thermal energy received at Earth's surface from the sun. Greenhouse-gas theory explains climate change by modulating thermal energy lost – ozone depletion theory explains climate change by modulating solar energy received (Fig. 2). There is much more thermal energy in play radiated by the sun than radiated by Earth.

Radiation transfers thermal energy from warmer matter, through air or space, to cooler matter. Thermal energy (heat) in matter is proportional to frequency as discussed below. In 1900, Planck⁹ had to postulate that thermal energy (**E**) in radiation is equal to frequency (**v**, the Greek letter nu) times the Planck constant (**h**), the number of joules (or electron volts) per cycle per second, in order to write an equation describing observed spectral radiance. **E=hv** is used in most photochemical

¹U.S. Geological Survey retired, Teton Tectonics, Jackson, Wyoming 83001, USA.

equations to signify the thermal energy required to cause a photochemical reaction typically driven by ultraviolet radiation. When the frequency (energy) is high enough, molecules are “shaken apart”, destroyed. $E=h\nu$ explains why infrared radiation does not have enough energy to be detected by our eyes but visible light does. $E=h\nu$ explains why visible light has enough energy to cause photosynthesis but infrared radiation does not. $E=h\nu$ explains why too much ultraviolet light would destroy any rods and cones sensitive to those frequencies in our eyes. As frequency (energy) increases to levels that damage DNA, cumulative length of exposure (dosage) becomes important. This is why extremely small doses of X-rays have enough energy to penetrate our bodies without harm, yet higher doses with the same energy can be focused to destroy cancer cells. Similarly for very high-frequency nuclear radiation, exposure above some minimum dosage causes illness and death. $E=h\nu$ is also the fundamental quantum of energy in modern physics.

$E=h\nu$ states that a narrow sliver of ultraviolet-B radiation received on Earth when ozone is depleted has frequency (energy) 48 times larger than the thermal energy of a much broader band of infrared radiation absorbed most strongly by carbon dioxide, because thermal energy is a function of frequency, not wavelength or bandwidth as currently assumed based on Maxwell’s wave equations¹⁰. Waves inhabit matter, not space or some luminiferous aether. Infrared radiation emitted from a pot of lukewarm water feels warm while ultraviolet-B radiation feels hotter than an oxy-acetylene torch. A torch warms Earth faster and to a higher temperature than a pot of lukewarm water. A little ultraviolet-B radiation burns our skin – something an ocean full of lukewarm water could never do.

Ozone is also depleted by chlorine and bromine gases emitted by explosive, effusive, and even quietly degassing volcanoes¹¹. Large explosive eruptions also eject megatons of sulphur dioxide into the lower stratosphere, forming an aerosol that reflects and scatters primarily the ultraviolet components of sunlight, causing net global cooling of $\sim 0.5^\circ\text{C}$ for three years (Fig. 2d). All other volcanic eruptions not explosive enough to form aerosols, deplete ozone, causing net global warming for up to one decade (Fig 2c).

Thermal energy is frequency. Heat (temperature) in matter results from very high-frequency oscillations of all chemical bonds that hold matter together. At room temperature, these oscillations are on the order of 30 terahertz (30 million million cycles per second). These very high frequencies reflect the very small atomic dimensions of the oscillators. Each bond oscillates around a minimum in potential energy between electrostatic forces that attract oppositely charged atomic and molecular components when they are near and electrostatic forces that repel similarly charged atomic and molecular components when they are too near.

Each chemical bond oscillates at some frequency over some amplitude determined by the strength of the bond and how much the bond expands and contracts as a function of temperature. Heating bonds increases both frequencies and amplitudes – increasing volume, decreasing density. Because all molecules in matter touch contiguous molecules, these frequencies and amplitudes can flow mechanically via conduction, becoming homogenised over some portion of matter, reaching thermal equilibrium where heat no longer flows. Heat flows spontaneously from hot to cold, from higher frequency and amplitude to lower frequency and amplitude. The rate of flow is proportional to the differences.

In 1900, Max Planck⁹ formulated empirically an equation (Planck’s law) describing the observed spectral radiance, the amount of radiation (the brightness) measured at each frequency as a function of the absolute temperature of matter. His equation assumes matter is a black body, a perfect absorber and emitter of radiation, whose surface is at thermal equilibrium. While these curves represent spectral radiance just above the surface, they must also represent the frequency and amplitude of oscillations on the surface. The motion of charge is well observed to generate an

electric field that induces a magnetic field, that induces an electric field, ad infinitum, forming electromagnetic radiation. This is how radio signals are transmitted.

The solid curves in Fig. 3 show spectral radiance emitted from bodies at various temperatures. Note (1) temperature of matter, as we perceive it, is a function of a broad spectrum of oscillations, (2) the width of this spectrum increases with increasing temperature, (3) spectral radiance (amplitude) at each frequency is determined by the temperature of the emitting body and increases with increasing temperature, (4) the frequency of the peak spectral radiance increases rapidly with temperature, (5) the broad spectrum of radiation from the surface of a body at one temperature does not overlap, at any frequency, the broad spectrum of radiation from the surface of another body at any other temperature, and (6) the broad spectrums do overlap when the bodies are at different distances so that their amplitudes (spectral radiance) are reduced by one over the square of the distance travelled.

Note that a body can only be warmed by radiation from a warmer body that contains higher frequencies and higher amplitudes. The dashed black line shows Wien's displacement law, the frequency (ν) of peak spectral radiance as a function of a body's temperature (T) where $\nu = 5.88 \times 10^{10} T$. Radiation consisting of a narrow band of frequencies close to this value or larger could warm a body to temperature T if absorbed in sufficient quantity. Radiation with peak spectral radiance less than ν can only warm a body to a lower temperature. This means that it is physically impossible for Earth to be warmed by its own radiation as assumed, for example, in energy budget calculations for greenhouse gases¹². Ultraviolet-B radiation that reaches Earth when ozone is depleted has the potential to warm Earth to 16,500K, but luckily only very small amounts are available and only during daytime.

Radiation just above the emitting surface consists of frequencies of oscillation with the amplitude at each frequency equal to the amplitude of oscillation on the surface of the emitting body. While conduction homogenises frequency and amplitude within matter to form a Planck distribution, in air or space, frequency and amplitude are completely independent. Frequency (colour) does not change even over galactic distances, except for Doppler effects, while amplitude decreases as the inverse square of the distance travelled because amplitude becomes spread out over a surface area that increases with the square of the distance. For radiation, amplitude is the brightness of the light that we see, the radiance, and it is set by the amplitude of oscillations on the surface of the emitting body.

In the 19th century, spectral radiance was measured by using a prism to separate frequencies spatially in order to measure the amount of radiation in each narrow frequency band using a thermocouple, a thermopile, or a temperature sensitive resistor. These measurements did not measure radiation directly – they measured the change in temperature of matter caused by radiation. This is why spectral radiance was thought of in terms of watts per square meter. This may be appropriate once radiation has been absorbed by matter. But for radiation in space or air shown by Planck's law, frequencies on the x-axis are proportional to energies (watts) ($E=h\nu$) and do not change with distance while amplitudes on the y-axis decrease with the square of the distance and do not contain units of energy or power (watts). Unfortunately all terms we have describing the amount (amplitude) of radiation include units of power (watts): spectral radiance, brightness, intensity, flux. Even luminous intensity measured in candelas is thought of as watts. Spectral radiance on the y-axis in Fig. 3 should not contain units of watts, which can be removed by replacing the Planck constant in Planck's empirical law with a constant without units of watts.

Ozone absorbs ultraviolet-B radiation. Ozone makes up only about 0.6 parts per million of the atmosphere by volume, but it is continuously created and destroyed in an endless photochemical Chapman cycle¹³, absorbing ultraviolet radiation to maintain an “ozone layer” between 15 and 35

km above Earth's surface. The stratosphere is heated daily primarily by continual dissociation of molecular oxygen (O_2) and ozone (O_3) where the molecular/atomic pieces fly apart at high velocity, increasing the average translational kinetic energy of all gas molecules, which, according to the ideal gas law, increases the temperature of air. The optical thickness of this layer varies regionally by the minute, time of day, season, and latitude⁷. Ozone tends to accumulate at the poles between mid-winter and early-spring when increasing sunlight forms ozone faster than it can be destroyed (Fig. 4)¹⁴. More ozone during winter heats the lower stratosphere, causing very cold temperatures on Earth. Ozone depletion since 1970 is most noticeable during these times with total column ozone decreasing ~50% each winter in the Antarctic ozone hole and less regularly in the Arctic but reaching 45% in 2011¹⁵. Less ozone cools the lower stratosphere and warms minimum surface temperatures. The greatest warming observed on Earth since 1970 was along the Antarctic Peninsula^{16,17}, central West Antarctica¹⁸, and in southern oceans¹⁹ in late winter, early spring, all within the Antarctic ozone hole. The second greatest warming was in the Arctic²⁰, well-known as Arctic amplification.

Between 1978 and 2008, ozone depletion increased ultraviolet-B radiation received on Earth²¹ 23% at 50°S, 9% at 35°S, 7% at 39°N, 9% at 50°N, decreasing temperatures in the lower stratosphere ~1°C^{22,23} (Fig. 5, purple line) and increasing Earth's mean surface temperatures ~0.6°C¹ (Fig. 1, red bars).

Substantial daily changes in ozone, especially north of 40°N, appear closely related to changes in atmospheric and surface temperatures, the depths and locations of surface pressure highs and lows, and the position of jet streams^{14,24} suggesting a link through ozone between long-term climate and short-term weather.

Ozone depletion and volcanoes. Chlorine and bromine gases emitted by volcanoes deplete ozone¹¹ with one atom destroying ~100,000 molecules of ozone because of the catalytic nature of the Chapman cycle⁴. Total column ozone, the amount of ozone in a vertical column above a point on Earth, varies rapidly⁷, but annual averages at each station tend to be similar. It is these averages that relate most directly to annual average surface temperatures.

The longest continuous measurements of total column ozone have been made since 1927 at Arosa, Switzerland (black line, Fig. 5)^{6,7}. The dashed grey line with blue data markers shows, for 1964-2009, the annual mean area-weighted total ozone deviation from 1964-1980 means in northern mid-latitudes (30-60°N) scaled from -8% at the bottom of the figure to 10% at the top²⁵. Years of increasing or decreasing ozone are nearly identical at Arosa and for this mean with small differences in amplitude. Thus the Arosa data provide a reasonable approximation for annual mean total column ozone throughout northern mid-latitudes since 1927.

Ozone at Arosa averaged 331 DU (Dobson units) until 1974, fell 9.4% to 300 DU by 1993 and began rising again until 2011. This long-term decrease has been reliably associated with an increase in anthropogenic tropospheric chlorine (green line, y-axis inverted) through chlorine catalysed destruction of ozone³ (Fig. 2b). Under the Montreal Protocol, production of CFCs was phased out, halting the increase in tropospheric chlorine by 1993.

Note the substantial drop in total column ozone the year following most volcanic eruptions (labelled in red). Ozone depletion is accompanied by decreasing temperatures in the lower stratosphere (purple line)^{22,23} especially following major explosive eruptions of Agung (1963), El Chichón (1982), and Pinatubo (1991)^{26,27}.

In 1991, the major eruption of Pinatubo depleted ozone 6% but exploded 17 megatons of sulphur dioxide into the lower stratosphere, forming a sulphuric-acid aerosol 15-25 km above Earth²⁸, spreading globally within months. The aerosol particles grew large enough to Rayleigh scatter the highest frequencies of sunlight (primarily ultraviolet-B), cooling Earth ~0.5°C for three

years (Fig. 2d). The ozone layer was most depleted during late winter, causing warming over continents²⁹, but the globally distributed aerosols caused net cooling (Fig. 2d). Such cooling has been observed after all major explosive volcanic eruptions throughout human history. The largest, Tambora in 1815, preceded the last cold phase of the Little Ice Age around 1850. When such eruptions happen frequently, their cumulative effects decrease ocean temperatures substantially^{30,31}, ultimately ushering in an ice age³².

The much smaller effusive eruptions of Eyjafjallajökull (2010) and Grímsvötn (2011) also depleted ozone ~6% each but did not eject sulphur dioxide into the stratosphere (Fig. 2c). Record warm temperatures and drought followed in North America during 2012-2013 related in time and location to 14% ozone depletion measured over Toronto Canada⁷: 3% due to CFCs and ~5.5% due to each of these small eruptions. Fig. 6 shows ozone concentrations over Toronto by month and year and compares winter-spring ozone depletion with minimum surface temperatures³³. Ocean temperatures east of New England reached the highest temperatures ever recorded³⁴ likely causing Great Britain's record flooding in 2012-2014. The previous major "Dust Bowl" drought in North America, which peaked in 1934-1936, was also preceded by a highly unusual sequence of seven small volcanic eruptions around the Pacific Ocean from 1931 through 1933.

The greatest warming at the end of the last ice age was contemporaneous with major, basaltic, effusive eruptions in Iceland³⁵ between 11,750 to 9,350 years ago³² (Extended Data Fig. 1).

Every 22 million years, on average³⁶, massive eruptions of basaltic lava covered areas as large as Australia or the contiguous United States. Some of these events erupted up to four million cubic kilometres of basaltic lava over hundreds of thousands of years, contemporaneous with lethally hot temperatures, acidic oceans, major ozone depletion, increased rates of mutation, and major mass extinctions^{37,38}.

Climate, throughout Earth's history, appears controlled by the frequency of major explosive volcanic eruptions causing ozone depletion but forming aerosols that caused net cooling (Fig. 2d) and the duration of effusive, basaltic volcanic eruptions not explosive enough to form aerosols but causing prolonged ozone depletion and associated warming (Fig. 2c). The relative proportion of these two types of volcanism is determined by plate tectonics processes that control the relative motion of tectonic plates. For example, 56.1 million years ago, global temperatures rose 6°C in less than 20,000 years associated with voluminous basaltic eruptions during the initial opening of the Greenland-Norwegian Sea³⁹.

Ozone depletion caused by volcanoes and CFCs appears to explain qualitatively most observed global warming. It will take considerable data gathering and modelling to determine quantitatively whether ozone depletion explains all global warming.

What actions should we take? We humans caused the world to warm ~0.6°C from 1970 to 1998. Heat is accumulating in the ocean⁸, resetting Earth's thermostat. The only natural way to lower the thermostat is through increased numbers of major explosive volcanic eruptions.

We have halted the increase in human-caused global warming by reducing emissions of CFCs via the Montreal Protocol. We need to strengthen the prohibition against manufacturing gases that deplete ozone and to seek ways to remove these gases from the atmosphere.

We also need to understand better exactly how volcanoes deplete ozone and what our options will be if the rates and types of global volcanism suddenly change.

Reducing emissions of carbon dioxide is not likely to reduce global warming but may slow ocean acidification.

Rapid increases in pollutants such as black carbon formed by incomplete combustion, sulphur dioxide from burning fossil fuels, and ground-level ozone formed as a result of nitrogen oxides, volatile organic compounds, and other pollutants occurred 30 years prior to major warming

(Extended Data Figs. 2, 3) suggesting that they do not have much effect on global warming other than absorbing some ultraviolet-B radiation. But these pollutants do have major deleterious effects on public health and many cause acid rain. Most developed countries have reduced pollution to relatively healthy levels, but improvements are still possible and of value. Developing countries rapidly increasing energy use, such as China and India, need to put major emphasis on reducing pollution for the health of their citizens and others living downwind. Concentrations of corrosive sulphate in acid-rains that reach the western United States from Asia and from ships at sea approach and sometimes exceed US maximum allowable levels⁴⁰.

The ozone depletion theory of global warming is described in detail at www.ozonedepletiontheory.info. Join the discussion of the science at [google discussion group to be set up].

[Reviewers can preview the website by going to www.ozonedepletiontheory.info/ozonemain/ with user name “jackson” and password “wyoming”.]

Online Content Any additional Methods, Extended Data display items and Source Data are available in the online version of the paper; references unique to these sections appear only in the online paper.

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Supplementary Information is available in the online version of the paper.

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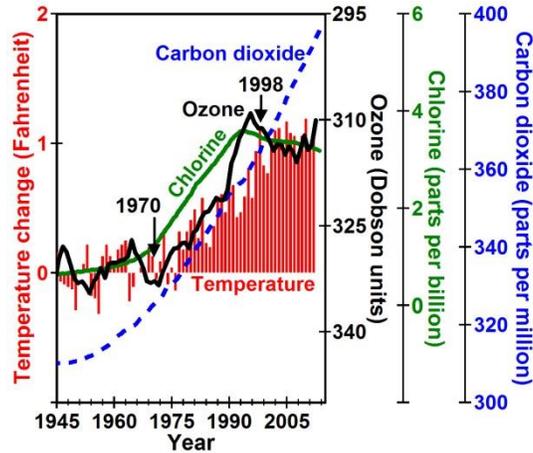


Figure 1 | The trend of carbon dioxide diverges from trends in temperature and ozone depletion. Trends in temperature (red bars), anthropogenic tropospheric chlorine (green line) and ozone (black line) over the past 70 years are distinctly different from trends in concentrations of greenhouse gases such as carbon dioxide (blue dashed line). Increases of chlorine concentrations in the troposphere tend to precede by 3 to 5 years increases in the stratosphere that deplete ozone³.

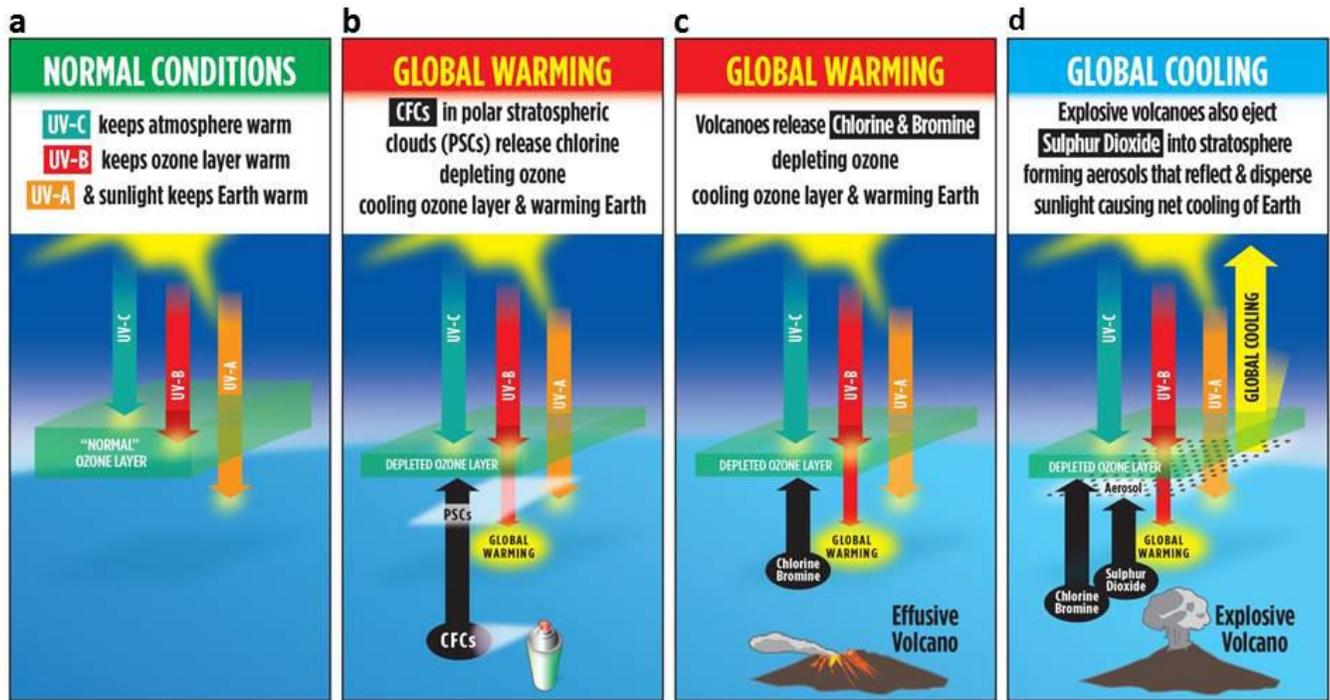


Figure 2 | The effects of ozone depletion and volcanic aerosols on climate. When the ozone layer is depleted either by human-manufactured CFCs (b) or by chlorine and bromine emitted by volcanic eruptions (c), more ultraviolet-B sunlight than normal reaches the ocean, warming Earth. When large, explosive volcanic eruptions eject megatons of sulphur dioxide into the lower stratosphere forming sulphuric-acid aerosols that reflect and scatter primarily ultraviolet-B components of sunlight, thermal energy reaching Earth is decreased, causing global cooling (d).

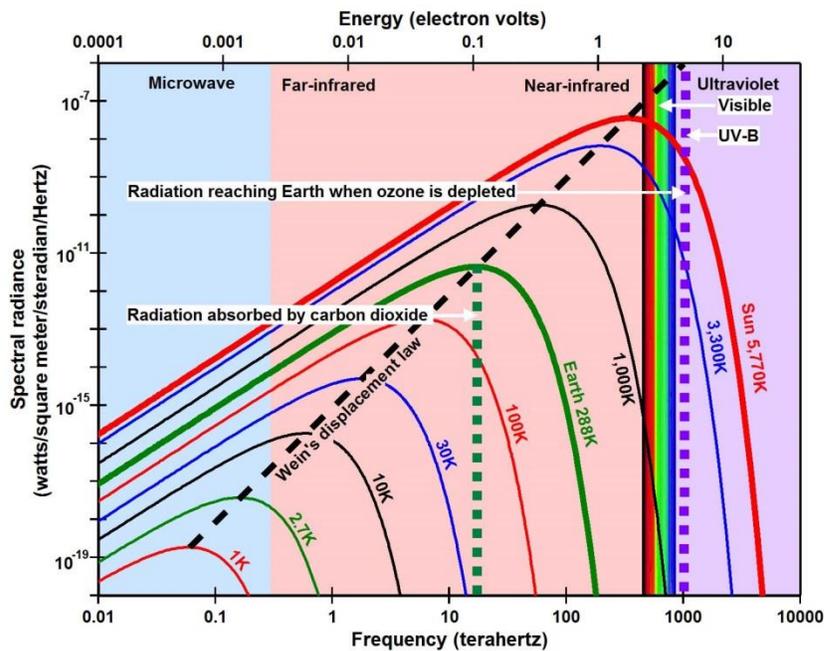


Figure 3 | Planck's law for spectral radiance, the amount of radiation at each frequency. These curves show the spectral radiance for bodies of matter at different absolute temperatures. The dashed black line shows the peak spectral radiance for each temperature and can be thought of as the temperature of the radiation. A body can only be warmed by radiation with higher peak spectral radiance. The dashed green line shows the peak spectral radiance for Earth (288K) absorbed by greenhouse gases such as carbon dioxide. The dashed purple line shows the peak spectral radiance for ultraviolet-B radiation (16,500K) that reaches Earth when ozone is depleted. Spectral radiance should not include units of watts as explained in the text.

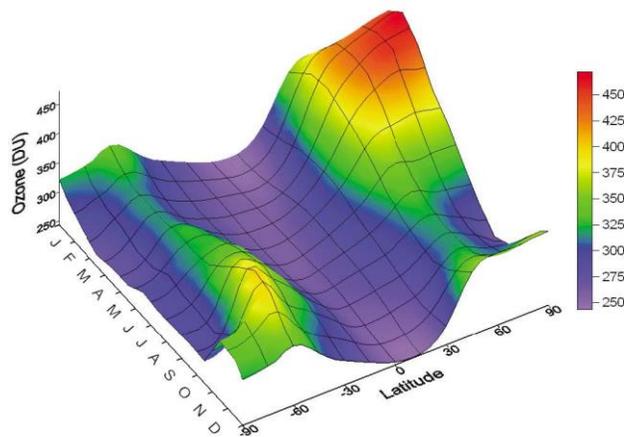


Figure 4 | Ozone accumulates during the winter in polar regions. Zonal monthly mean total column ozone in Dobson Units shown as a function of latitude and month estimated from ground-based data for the period 1964 to 1980¹⁴. Excess ozone means more solar ultraviolet-B radiation is absorbed in the lower stratosphere, causing temperatures to become particularly cold near the ground. Ozone depletion is greatest during these late winter to early spring peaks.

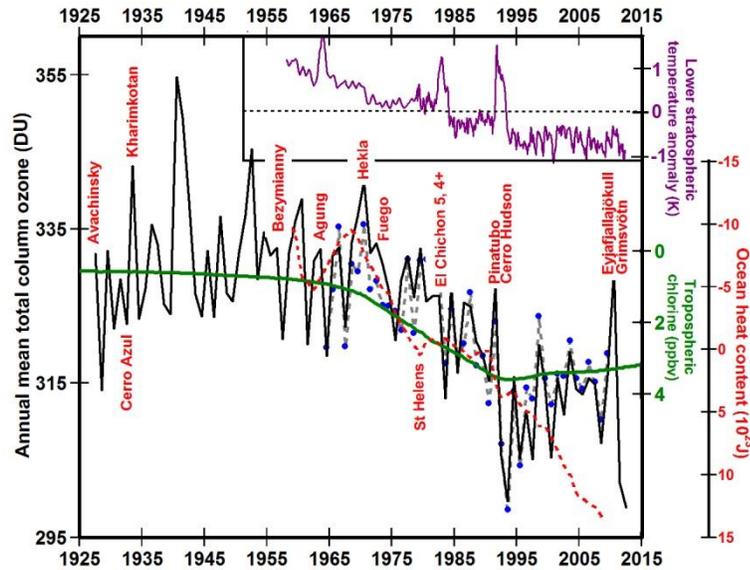


Figure 5 | Ozone concentration decreases in the year following most volcanic eruptions. Annual mean total column ozone (black line)^{6,7} decreases ~6% during the year following volcanic eruptions, cooling the lower stratosphere (purple line)^{22,23} and warming the ocean (dashed red line, y-axis inverted)⁸. Names of the erupting volcanoes are shown in red. The green line shows annual mean tropospheric anthropogenic chlorine with the y-axis inverted³ and has been scaled to have a slope similar to the slope for ozone between 1970 and 1991. This figure is described in detail at ozonedepletiontheory.info/depletion.html.

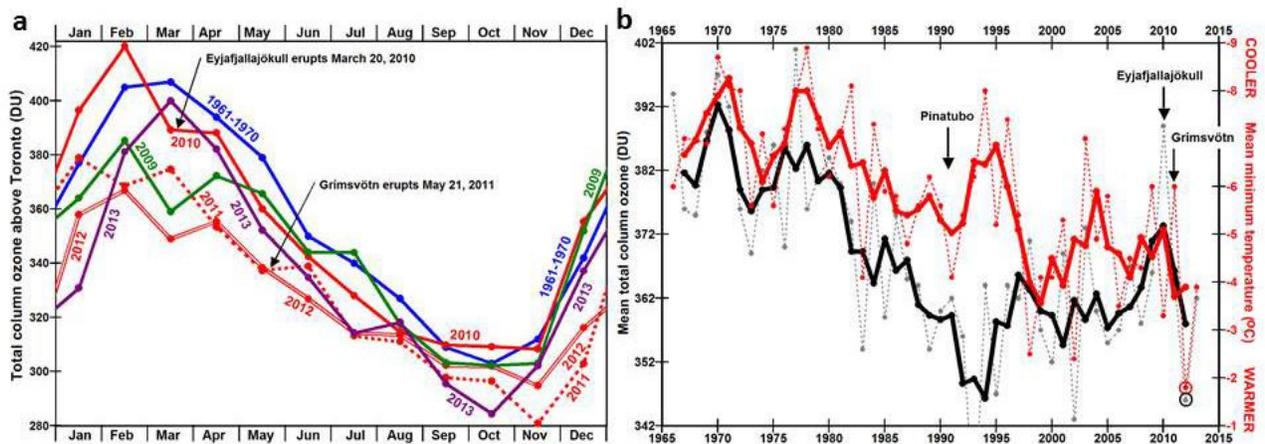
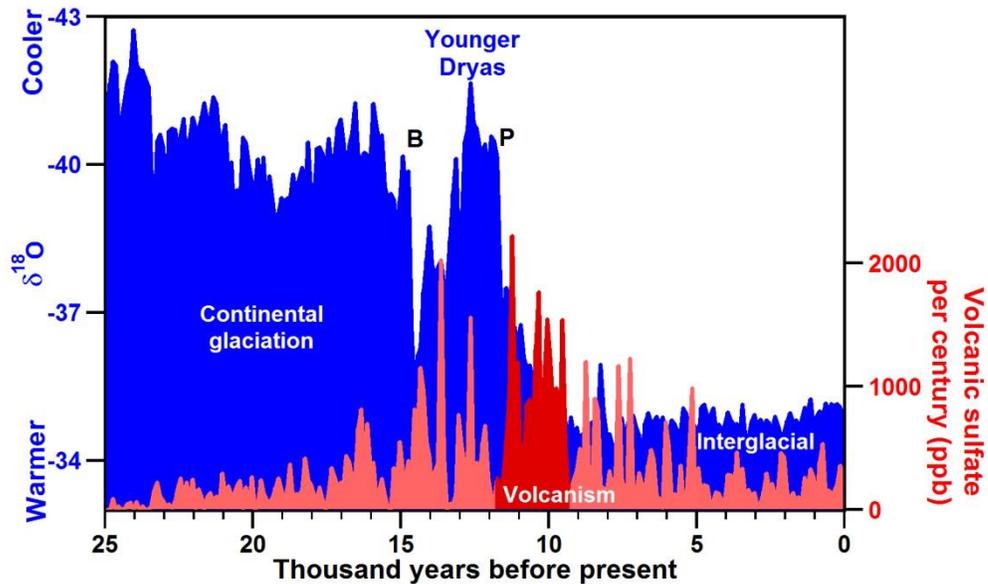


Figure 6 | Ozone concentrations above Toronto, Canada, reached a minimum in 2012 when minimum temperatures reached a maximum. **a**, Total column ozone⁷ per month in November, 2011 (dotted red line), was 12% below the average for Novembers in 1961 through 1970 (blue line) and has remained unusually low throughout 2012. **b**, When mean total column ozone measured during the months of December through April (black line) decreased, mean minimum temperature³³ for the same months typically warmed (red lines, y-axis inverted) except following the eruption of Pinatubo in 1991. The dashed lines show annual means; the solid lines are smoothed using a 3-point centered running mean.



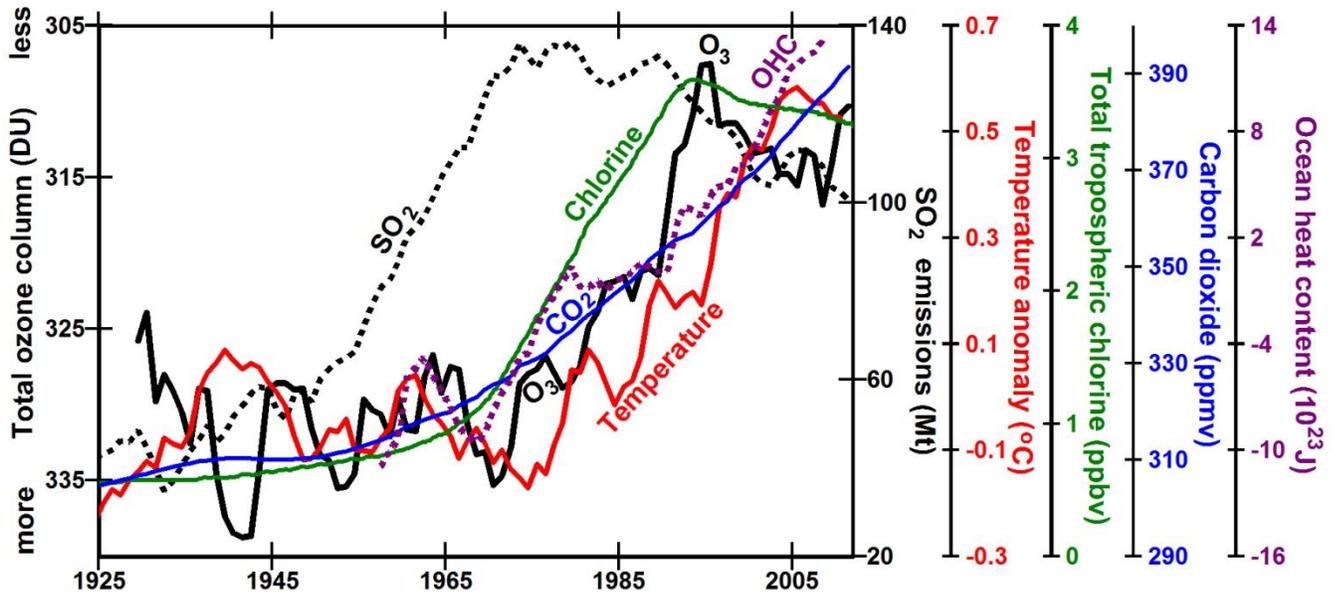
Extended Data Figure 1 | Volcanism was highest and most continuous precisely as the world was warmed out of the last ice age. Volcanic sulfate (red) measured in ice below Summit Greenland shows a peak in volcanism between 11,750 and 9,350 years ago (darker red), the ages of many basaltic lavas in Iceland. This is the same time when the world warmed out of the last ice age according to an oxygen isotope proxy for local temperature (blue) measured in the same ice. B is the Bolling warming and P is the Pre-boreal warming.

Data:

Volcanic sulfate: <ftp.ncdc.noaa.gov/pub/data/paleo/icecore/greenland/summit/gisp2/chem/volcano.txt>

Oxygen isotopes: <ftp.ncdc.noaa.gov/pub/data/paleo/icecore/greenland/summit/gisp2/isotopes/stacked.txt>

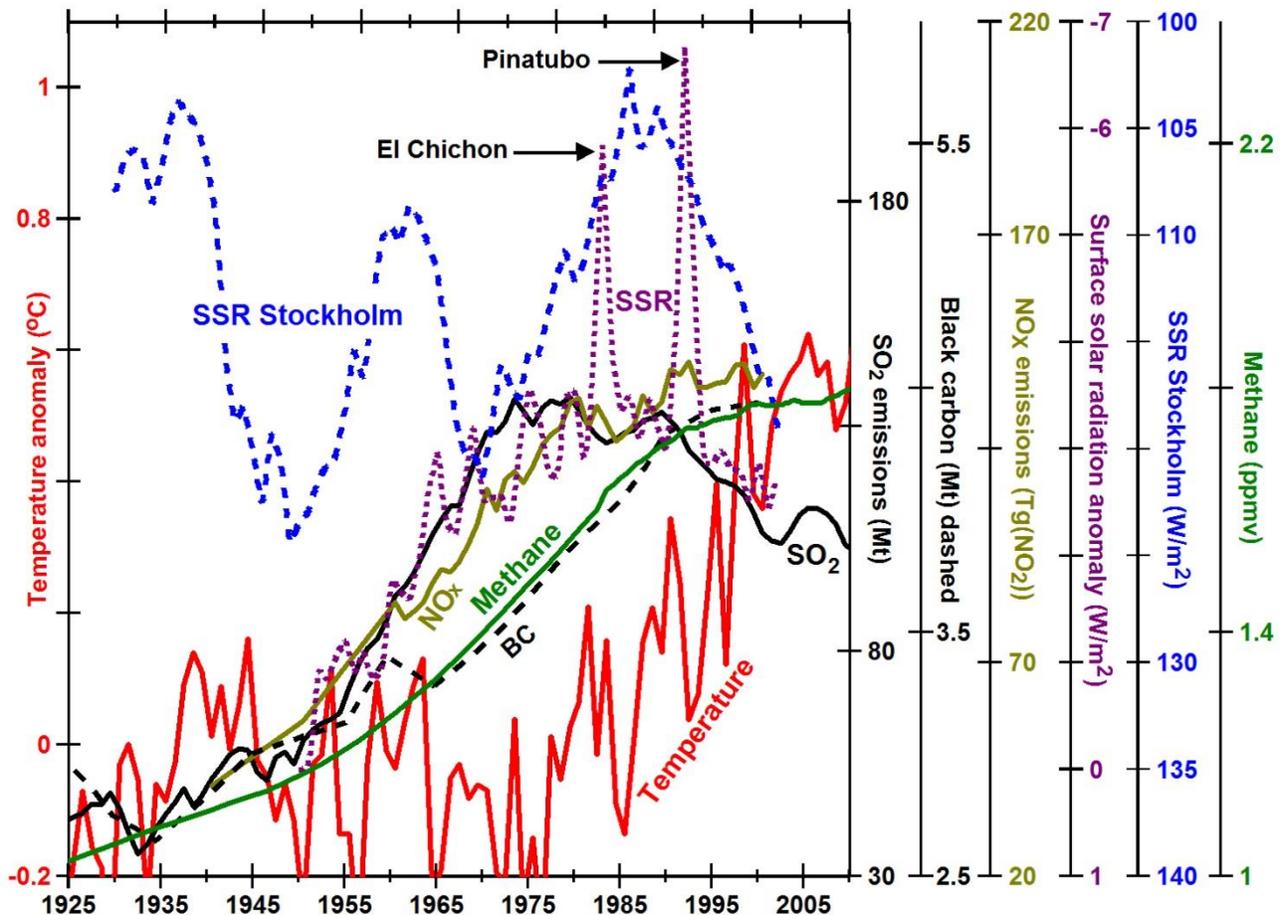
Volcanism in Iceland: Licciardi, J. M., Kurz, M. D. & Curtice, J. M. Glacial and volcanic history of Icelandic table mountains from cosmogenic ³He exposure ages. *Quat. Sci. Rev.* 26, 1529-1546 (2007).



Extended Data Figure 2 | Increased SO₂ pollution occurred too early to have had a major effect on global warming. SO₂ pollution (dotted black line) increases ~30 years before global warming (red line), ~20 years before anthropogenic tropospheric chlorine (green line) increased, and ~25 years before total column ozone decreased (black line, y-axis inverted). Mean annual temperature anomaly in the Northern Hemisphere (red line) and ozone (black line) are smoothed with a centered 5 point running mean. OHC is ocean heat content (dotted purple line).

Data:

- Ozone:** Staehelin, J. *et al.* Total ozone series at Arosa (Switzerland): Homogenization and data comparison. *J. Geophys. Res.* **103**, 5827-5841 (1998) and WOUDC. *World Ozone and Ultraviolet Radiation Data Center*, <www.woudc.org/data_e.html> (2014).
- SO₂:** Smith, S. J. *et al.* Anthropogenic sulfur dioxide emissions: 1850–2005. *Atmos. Chem. Phys.* **11**, 1101-1116 (2011).
- Temperature:** www.cru.uea.ac.uk/cru/data/temperature/HadCRUT4-nh.dat
- Chlorine:** Solomon, S. Stratospheric ozone depletion: A review of concepts and history. *Rev. Geophys.* **37**, 275-316 (1999), Plate 6.
- Carbon dioxide:** www.esrl.noaa.gov/gmd/ccgg/trends/#mlo_data and cdiac.ornl.gov/ftp/trends/co2/lawdome.combined.dat
- Ocean heat content:** Levitus, S. *et al.* World ocean heat content and thermosteric sea level change (0-2000 m), 1955-2010. *Geophys. Res. Lett.* **39**, L10603 (2012), Figure 1.



Extended Data Figure 3 | There is an unexplained time lag between global warming and increases in most pollutants and greenhouse gases. Trends in SO_2 , NO_x , black carbon, and methane pollution increase 20 to 30 years before mean global surface temperatures. The dotted purple line shows the decrease in surface solar radiation (SSR) predicted by modelling these pollution sources. The dashed blue line shows surface solar radiation observed in Stockholm, Sweden. These pollutants do not appear to have much effect on solar energy reaching Earth and they occur too early to have directly affected mean global surface temperatures.

Data:

Temperature: www.cru.uea.ac.uk/cru/data/temperature/HadCRUT4-nh.dat

SO_2 : Smith, S. J. et al. Anthropogenic sulfur dioxide emissions: 1850–2005. *Atmos. Chem. Phys.* **11**, 1101–1116 (2011).

Black carbon: Bond, T. C. et al. Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000. *Global Biogeochem. Cycles* **21**, GB2018 (2007). The drop in 1965 reflects improvements in diesel-engine design first applied in this accounting in 1965.

NO_x : EPA. *Air emission sources*, <www.epa.gov/air/emissions/> (2014) in United States only.

SSR: Wild, M. Global dimming and brightening: A review. *J. Geophys. Res.* **114**, D00D16 (2009) Figure 5.

SSR Stockholm: Wild, M. Global dimming and brightening: A review. *J. Geophys. Res.* **114**, D00D16 (2009) Figure 2.

Methane: Etheridge, D. M., Steele, L. P., Francey, R. J. & Langenfelds, R. L. Atmospheric methane between 1000 A.D. and present: Evidence of anthropogenic emissions and climatic variability *J. Geophys. Res.* **103**, 15979–15993 (1998) and Dlugokencky, E. J. et al. Observational constraints on recent increases in the atmospheric CH_4 burden. *Geophys. Res. Lett.* **36**, L18803 (2009).

Chlorine: Solomon, S. Stratospheric ozone depletion: A review of concepts and history. *Rev. Geophys.* **37**, 275–316 (1999).