Rates of change in natural and anthropogenic radiative forcing over the past 20,000 years

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The rate of change of climate codetermines the global warming impacts on natural and socioeconomic systems and their capabilities to adapt. Establishing past rates of climate change from temperature proxy data remains difficult given their limited spatiotemporal resolution. In contrast, past greenhouse gas radiative forcing, causing climate to change, is well known from ice cores. We compare rates of change of anthropogenic forcing with rates of natural greenhouse gas forcing since the Last Glacial Maximum and of solar and volcanic forcing of the last millennium. The smoothing of atmospheric variations by the enclosure process of air into ice is computed with a firn diffusion and enclosure model. The 20th century increase in CO_2 and its radiative forcing occurred more than an order of magnitude faster than any sustained change during the past 22,000 years. The average rate of increase in the radiative forcing not just from CO_2 but from the combination of CO_2, CH_4, and N_2O is larger during the Industrial Era than during any comparable period of at least the past 16,000 years. In addition, the decadal-to-century scale rate of change in anthropogenic forcing is unusually high in the context of the natural forcing variations (solar and volcanoes) of the past millennium. Our analysis implies that global climate change, which is anthropogenic in origin, is progressing at a speed that is unprecedented at least during the last 22,000 years.

climate change | global warming | greenhouse gas | ice core

Measurements on atmospheric air samples and on air from ice and firn cores reveal an exceptional rise in the concentrations of the anthropogenic greenhouse gases carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) over the past two centuries (1) (Fig. 1). Today's concentration of these greenhouse gases are higher than ever measured over the last 650 thousand years (ka), the period covered by the ice core record (2, 3). The recent rise is man-made and causes a perturbation in the radiative balance of the planet (4) leading to global warming. Concerns about the impacts and costs of anthropogenic climate change have led to legally binding agreements to avoid dangerous anthropogenic climate interference (United Nation Framework Convention on Climate Change, 1992).

An important aspect of anthropogenic climate change is its rate of change. It codetermines the magnitude and severity of the impacts on socioeconomic and natural systems. A slow rate of change in forcing and resulting climate permits more time for adaptation than a fast rate of change. However, it remains difficult to quantify rates of past global temperature change because proxy data are of limited spatiotemporal resolution (5). On the other hand, past forcing from well mixed greenhouse gases can be accurately established from ice core data. Although today's unusually high greenhouse gas concentrations are widely discussed, less attention has been paid to the rate of change in their concentrations (6, 7) and in their radiative influence (8). Here, we quantify by how much the rate of change in greenhouse gas concentrations and their radiative forcing is accelerating. We address how current rates of increase compare with past rates as recorded in the ice core records.

The effects of the enclosure process of atmospheric air into ice as well as sample frequency must be taken into account to quantify rates. Before air is enclosed in ice, it enters the porous firn column that is overlying the ice with a thickness of 80–120 m. This leads to two important effects: (*i*) the age difference between the air entrapped in ice and the surrounding ice and (*ii*) the age distribution of gas in the air bubbles within one sample. The first effect is taken into account during the construction of the age scales for the ice core. The second effect causes a smoothing of atmospheric variations. In the firn, the air exchanges with the overlying atmosphere through the open pore system by molecular diffusion (9). Therefore, the air isolated in bubbles has not a discrete age, but an age distribution. In addition, and more important at sites with a low accumulation rate, most bubbles are formed at the transition from firn to ice over a depth interval of ≈15 m, which makes the age distribution even wider. Hence, fast variations in atmospheric trace gases are smoothed in the firn column and recorded in the bubbles as attenuated signals. The width of the age distribution (width at half peak-height) depends on the accumulation rate of snow. It varies from ≈20 years in cores from sites with a high accumulation rate (10–12) to up to ≈200 years for low accumulation rate sites such as Dome C, Antarctica. During glacial conditions, when accumulation rate and temperature are lowest, the age distribution in Antarctic cores can be as wide as ≈350 years.

Today, anthropogenic and natural factors exert changing radiative influences. The concept of radiative forcing is used to compare these (13). Anthropogenic factors are changes in the well mixed greenhouse gases CO_2, CH_4, and N_2O, in halocarbons and SF_6, in soot, tropospheric ozone (O_3), and stratospheric water vapor, all causing warming, aerosols, the surface albedo due to land use, and stratospheric O_3 (globally causing cooling). Natural forcing factors include changes in stratospheric sulfate loading caused by explosive volcanism (14) and changes in solar energy output (15, 16).

The goals of this paper are to (*i*) discuss the recent anthropogenic rise in greenhouse gases in the context of the ice core records, (*ii*) analyze the rate of change in atmospheric greenhouse gas concentration as recorded in the ice core record, taking into account uncertainties arising from sampling frequency and smoothing occurring during the enclosure of air in firn and ice, and (*iii*) compare radiative forcing and the recent rate of change in radiative forcing by greenhouse gases and other forcing factors.

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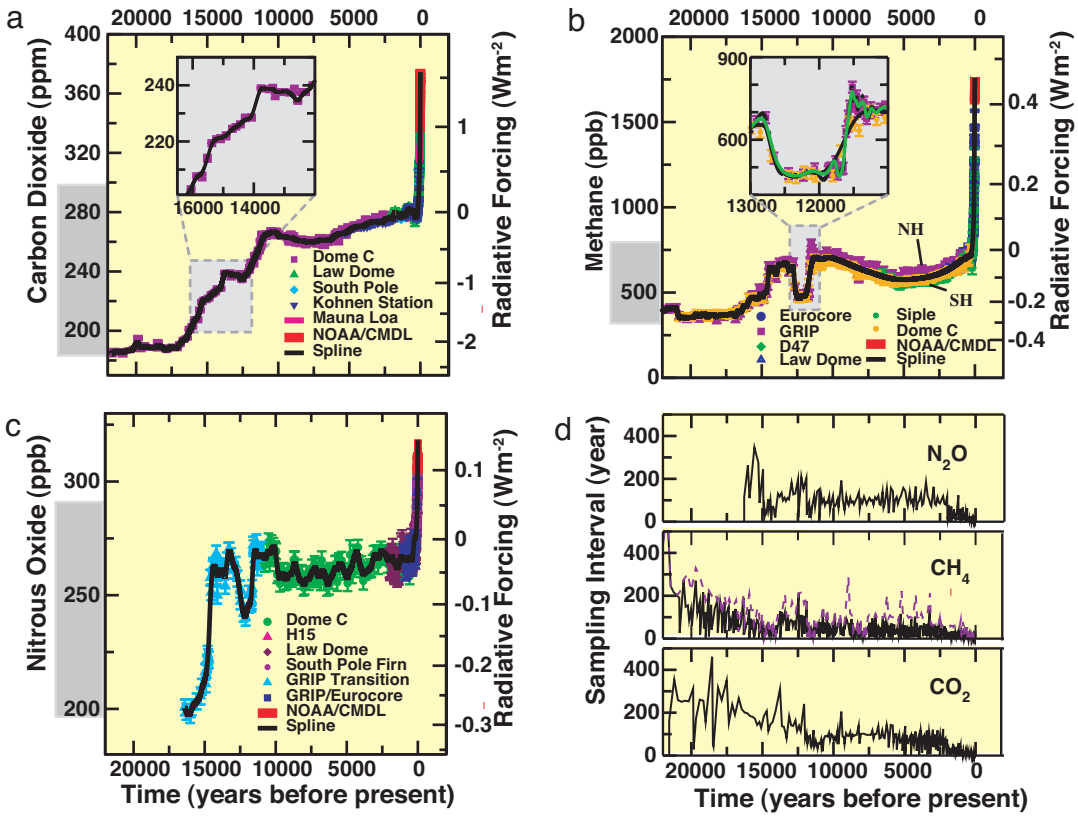

Fig. 1. Evolution of atmospheric CO_2 (*a*), methane (*b*), and nitrous oxide (*c*), and sampling intervals (*d*) over the past 20,000 years. The gray bar denotes the range of the preindustrial, natural variability in the concentrations of CO_2, CH_4, and N_2O as measured for the past 650,000 years. The green line in *b Inset* is a spline to the Greenland CH_4 data only that preserves multidecadal variability. Sampling intervals for the Greenland CH_4 data are shown by the dashed line in *d*. Data sources are given in *SI Text*.

Results

Greenhouse Gas Concentrations and Radiative Forcing. Atmospheric concentrations of CO_2, CH_4, and N_2O, as measured in the ice cores, have varied within restricted ranges over the past 650 ka before the Industrial Era (Fig. 1) (2, 3). CO_2 concentrations varied naturally between the lowest glacial values of 180 ppm and interglacial values of up to 300 ppm. Natural CH_4 variations were between 320 and 790 ppb, and natural N_2O variations between 195 and 290 ppb over the last 650 ka. Within the last 200 years, the maximum of the late Quaternary natural range has been exceeded by at least 25% for CO_2, 120% for CH_4, and 9% for N_2O. All three records show effects of the large and increasing growth in anthropogenic emissions during the Industrial Era.

Sample frequency [Fig. 1*d* and supporting information (SI) Fig. 5*d*) in the ice core is generally high enough to capture multidecadal to century scale variations over the past 22 ka and to record multidecadal variations over the past millennium. Many samples have been analyzed around periods of large variations, such as the transition to the Bølling, the end of the Younger Dryas, and the 8.2 ka event, when data spacing is sometimes as short as 30 years (SI Fig. 6).

A gas diffusion and enclosure model (17) is used to calculate the width of the age distribution for the different trace gases and ice cores and the attenuation of atmospheric signals during the enclosure process. The width of the age distribution is ≈200 years in the Antarctic data for the last transition, whereas the CH_4 data from Greenland are from samples with a small width of the age distribution (20–25 year) (SI Fig. 7). The comparison between the two hemispheric data sets reveals that decadal-to-century scale concentration changes are recorded also in Antarctic cores, at least over the past 22 ka (Fig. 1*b* and SI Fig. 6).

Using the available data and considering the smoothing effect of the ice archive, the data resolution is sufficient to exclude with very high confidence a peak similar to the anthropogenic rise for the past 50,000 years for CO_2, for the past 80,000 years for CH_4, and for the past 16,000 years for N_2O (*SI Text*).

Until today, the Industrial Era increase in CO_2, and in the radiative forcing by all three gases, is similar in magnitude to the increase over the transitions from glacial to interglacial periods, but started from an interglacial level and occurred much faster (Fig. 1). Radiative forcing from the three greenhouse gases increased by 2.3 W m^{-2} over the 6,000 years of the last glacial–interglacial transition, by 0.3 W m^{-2} over the Holocene, and by 2.2 W m^{-2} from 1750 to 2004 AD.

Variations in atmospheric CO_2 dominate the radiative forcing by all three gases over the industrial period and glacial interglacial cycles (Fig. 1). Radiative forcing by CO_2 increased by 3.8 W m^{-2} since the Last Glacial Maximum, but only by 0.6 and 0.4 W m^{-2} for CH_4 and N_2O. Consequently, most emphasis must be given to variations in CO_2 when reconstructing rates of change in forcing.

Rates of Change. We start our discussion of rates of change by determining average rates for distinct periods of the last 20 ka using the simplest approach. The records are divided into 8 and 11 distinct periods of different lengths for CO_2 and CH_4, respectively. Periods are selected such that the rate of change is approximately steady during each period. An average rate is determined by subtracting the measurement at the end from that

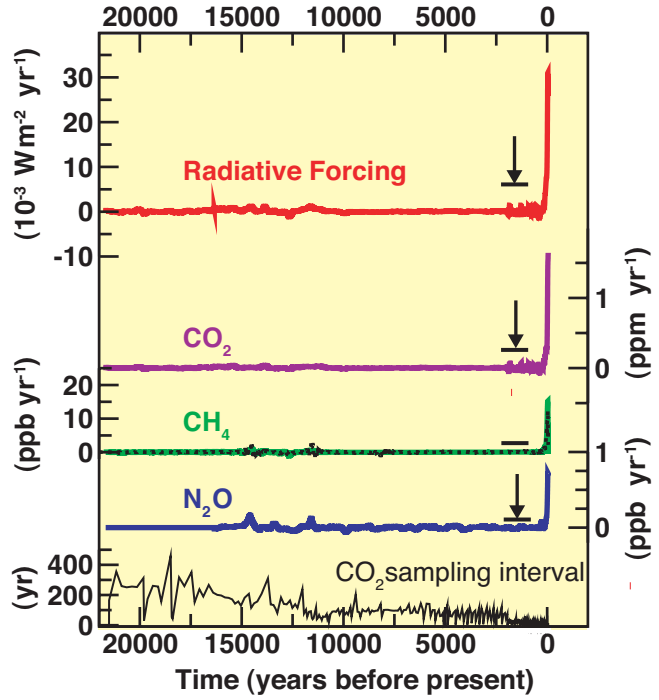

Fig. 2. Rates of change for the three greenhouse gases, N_2O, CH_4, CO_2 and their combined radiative forcing for the last 22 ka. The Greenland CH_4 data represent decadal-scale variations and inferred rates of change (dash) are directly comparable for different periods. The black lines/arrows show the peak in the rate of change in radiative forcing and concentrations after the anthropogenic signals of CO_2, CH_4 (no arrow shown), and N_2O have been smoothed with a model describing the enclosure process of air in ice (17) (*SI Text*).

at the beginning of each period and by dividing by the length of the period (*SI Text*). Selected periods are longer than the width of the age distribution. Thus, the effects of smoothing on the computed average rates are minimized.

For CO_2, the greatest average rate before the Industrial Era is 3.6 ppm/century from 14.6 ka BP to 14.3 ka BP (SI Table 1). This is 20 times smaller than the average rate of 71 ppm/century found for the 20th century. The largest increase over any individual preindustrial period of 31 ppm occurred over 1,600 years. For comparison, CO_2 rose by 31 ppm in just the last 20 years. For CH_4, the greatest preindustrial rate is 146 ppb/century found during the 170-year period starting at 11.7 ka BP (SI Table 2). This is six times smaller than the 20th century rate of 888 ppb/century. Correspondingly, the largest rate in CO_2 radiative forcing found during the preindustrial period is 0.083 W m^{-2}/century, 14 times smaller than the 20th-century rate of 1.16 W m^{-2}/century.

Next, we compute century-scale rates of change in a continuous way for the past 22 ka from spline fits (18) (*SI Text*) to the concentration data (Fig. 2). We assume that atmospheric N_2O has not varied during the period 16–22 ka BP, for which no N_2O data are available. The inferred rates of change in the concentrations of the three greenhouse gases and their combined radiative forcing have been higher during the industrial period than for any other period during the past 22 ka. The recent rate of increase in radiative forcing is $\approx 30 \times 10^{-3}$ W m^{-2} yr^{-1}, whereas inferred preindustrial rates are below 2×10^{-3} W m^{-2} yr^{-1}.

How do results depend on the details of the spline fitting procedure? This is illustrated by comparing the splines to the global CH_4 record and to the Northern Hemisphere record with a spline that follows the Northern Hemisphere data very tightly. The latter high-frequency spline has been produced by dividing the cut-off period used in the standard Northern Hemisphere CH_4 spline by a factor of two. This corresponds to a very extreme choice as the cut-off period is now equal or smaller than the data spacing. Deduced rates of change for the preindustrial period are in the range of -1.3 to 3 ppb yr^{-1} for the standard spline to the NH data, and of -4 to 4 ppb yr^{-1} for the high-frequency spline to the NH data. In any case, these rates are smaller than the average rate of increase over the 20th century of 8.8 ppb yr^{-1}. Thus, numerical results do somewhat depend on the details of the fitting procedure, but differences remain limited, and our main conclusions do not depend on the choice of details.

Finally, we consider how the enclosure process of air into firn and ice attenuates peaks in concentrations and forcing and could therefore lead to an underestimation of the rates. This effect is potentially important for decadal-to-century scale variations in CO_2 and N_2O that are reconstructed from Antarctic data with a typical width of the age distribution of 200 years. On the other hand, the age width is 20–25 years in the Greenland cores and decadal-scale changes in CH_4 are well recorded. The anthropogenic signals of CO_2, CH_4, and N_2O are smoothed with our gas enclosure model (17) (*SI Text*); it is assumed that the concentration peaks decrease after 2004 immediately to zero for CH_4, according to an exponential decay for N_2O with a lifetime of 120 years, and through uptake by the land biosphere and the ocean as calculated with the Bern carbon cycle model (19). The smoothed rate of change for the anthropogenic peak in radiative forcing would be 6.1 10^{-3} W m^{-2} yr^{-1} (Fig. 2). This is at least three times larger than any reconstructed preindustrial rate. The comparison of the CH_4 evolution around the Younger Dryas period (Fig. 1*b Inset*) as recorded at Dome C, Antarctica (age width: ≈200 years) and in Greenland ice (age width: ≈25 years) provides further compelling evidence that sustained changes in greenhouse gas concentrations are recorded in the Dome C core.

We draw the following conclusions. (*i*) During the Industrial Era, the average rate of increase in the combined radiative forcing from CO_2, CH_4, and N_2O is larger than at any time during the past 16 ka. (*ii*) Decadal-scale growth rates in CH_4 as found during the 20th century are several times larger than decadal-scale rates during the past 22 ka. (*iii*) The 20th century increase in CO_2 and in its radiative forcing occurred more than an order of magnitude faster than any multicentury scale change during the past 22 ka.

Decadal-scale rates are computed for the past 2,000 years from the Law Dome cores and from direct atmospheric measurements. The smoothing of the atmospheric signal is small at Law Dome, a high accumulation rate site in Antarctica, spanning the past two millennia (11). Thus, the decadal-scale rates of change computed from the Law Dome data are directly comparable to the rates computed from atmospheric measurements. Fig. 3 shows the acceleration in the growth rate of the three greenhouse gases and their combined radiative forcing during the industrial period. The average rate of increase in atmospheric CO_2 was at least five times larger over the period from 1960 to 1999 than over any other 40-year period during the period 1–1800 AD. The average rate of increase in atmospheric CH_4 was at least six times larger, and that for N_2O at least two times larger over the past four decades, than at any time during the past 1,800 years before the Industrial Era. Correspondingly, the recent average rate of increase in the combined radiative forcing by all three greenhouse gases (28×10^{-3} W m^{-2} yr^{-1}) was at least six times larger than at any time during the period 1–1800 AD.

The growth rates of CO_2 and N_2O continue to increase up to present (2004 AD), whereas the rate of increase in atmospheric CH_4 peaked around 1980. The growth rate of CH_4 declined and CH_4 concentration remained approximately stable during the past few years. This decrease in the CH_4 growth rate is also

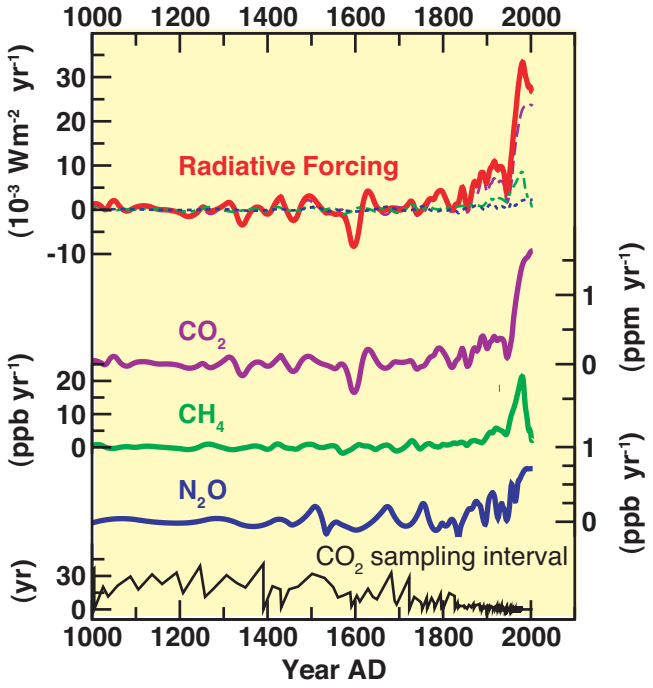

Fig. 3. Decadal-scale rates of change for the three greenhouse gases N_2O, CH_4, CO_2 and their combined radiative forcing over the past millennium. Rates were computed from the spline fits to the Law Dome (11) and atmospheric data shown in the main panels of SI Fig. 5. The thin lines at the top indicates the rate of change in forcing from CO_2 (violet, dash), CH_4 (green, dash-dot), and N_2O (blue, dot).

reflected by a slight decrease in the rate of the combined radiative forcing.

Discussion and Conclusion

Rates of change in greenhouse gas concentrations and in radiative forcing have been reconstructed for the past 20 ky. Uncertainties arise from the attenuation of atmospheric gas signals during the enclosure process of air into ice (17, 20, 21) and from sampling resolution. The results show that the 20th century rise in the concentration of CO_2 and CH_4 and in the combined radiative forcing from CO_2, CH_4, and N_2O is exceptionally high in the context of the past 20 ka.

Are the rates of anthropogenic forcing higher than those of natural forcing factors? We further analyze radiative forcing from solar irradiance changes and explosive volcanisms as well as from anthropogenic sulfate aerosols and from halocarbons and SF_6 to put the anthropogenic rise in forcing into the perspective of decadal-to-century scale natural forcing variability of the last millennium (Fig. 4; *SI Text* and SI Table 3). Changes in orbital parameters vary over multimillennial periods and were small for the last 1,000 years. Considerable uncertainties exist in the temporal evolution and the magnitude of solar irradiance changes, volcanic forcing, and tropospheric aerosol forcing (4). These uncertainties affect any comparison with the well defined 20th century greenhouse gas record.

The anthropogenic sulfur emission history of ref. 22 shows an increase over the industrial period with a peak in the late 1980s and a subsequent decrease by 24% until 2000 AD. The rate of change in (negative) sulfate aerosol forcing is estimated to increase from -23×10^{-3} W m^{-2} yr^{-1} around 1960 to $+15 \times 10^{-3}$ W m^{-2} yr^{-1} in year 2000 (Fig. 4*b*) with an average of $\approx 3 \times 10^{-3}$ W m^{-2} yr^{-1} over this period. The high and positive rates estimated for the past years may be biased high as the sulfate burden decreased less than sulfur emissions (23). Forcing

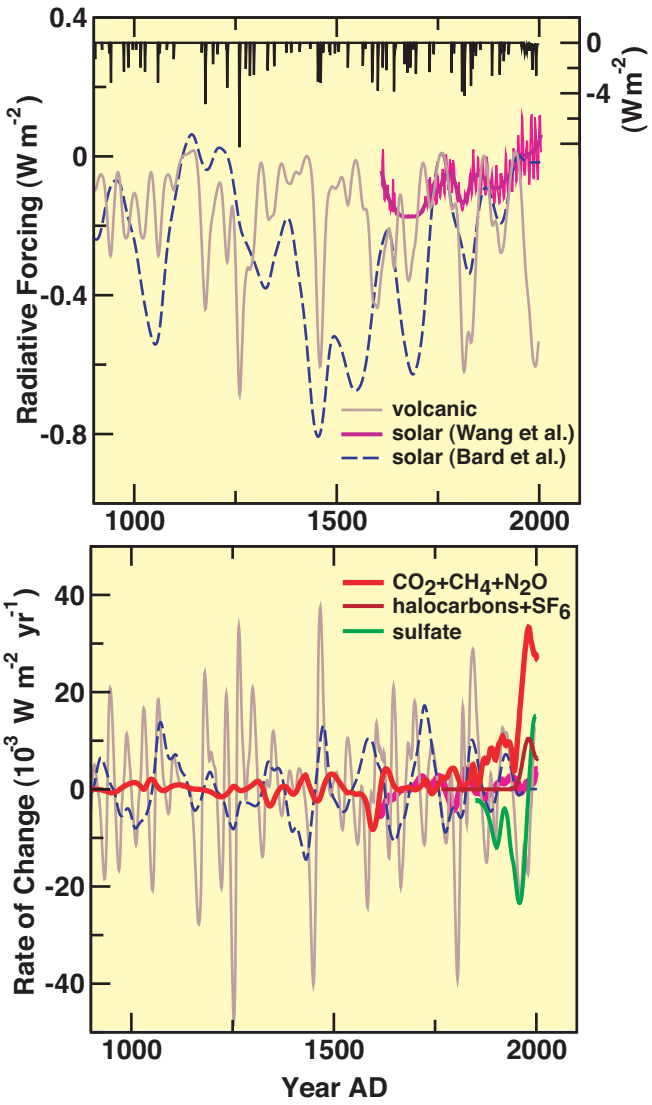

Fig. 4. Forcings and their rates of change during the last millenium. (*Upper*) Decadal-scale solar (15, 27) and volcanic (28) forcing during the last millennium expressed relative to 1750 AD. Data were splined with a 40-year cutoff period. The original reconstructions of volcanic forcing (28) (black) and of solar forcing from (15) (thin, magenta) are shown as well. (*Lower*) Rates of change in radiative forcing from explosive volcanism, solar changes, the three greenhouse gases (CO_2, CH_4, and N_2O), sulfate aerosols, halocarbons, and SF_6.

from halocarbons and SF_6 has also increased over the industrial period and the implied rate is $\approx 8 \times 10^{-3}$ W m^{-2} yr^{-1} over the past 40 years, with a very recent slowing of growth (Fig. 4*b*). We estimate that other anthropogenic forcing factors such as ozone, soot, and albedo changes contribute a few 10^{-3} W m^{-2} yr^{-1} to the recent trend (*SI Text*). Furthermore, the average rate of increase in anthropogenic forcing is estimated to total $\approx 35 \times 10^{-3}$ W m^{-2} yr^{-1} for the period 1960–2000. We conclude that the recent decrease in sulfate aerosol emissions as well as emissions of halocarbon and SF_6 contribute substantially to the current growth trend in anthropogenic forcing.

Turning to natural forcings, there is an ongoing debate on the importance of solar forcing. The latest summaries of the various uncertainties can be found in two recent reviews (16, 24). Model-based analyses comparing results from simulations with different magnitudes of solar forcing and the Northern Hemisphere temperature and atmospheric CO_2 proxy records suggest

a limited role of solar changes for last millennium climate variability (25, 26). Here, we use the solar forcing reconstruction of Bard *et al.* (27) and Wang *et al.* (15) (Fig. 4). The Bard *et al.* reconstruction is based on the assumption that solar irradiance was reduced by 0.25% relative to present during the Maunder Minimum), whereas the more recent work by (15) yields a Maunder Minimum reduction of ≈0.1%. During the past millennium, the rates of change in solar forcing vary between −15 and + 17 10^{-3} W m^{-2} yr^{-2} for the Bard *et al.* reconstruction and between −6 and +4 × 10^{-3} W m^{-2} yr^{-1} for the Wang *et al.* reconstruction. In any case, these rates are smaller than the rates in recent greenhouse gas forcing (Fig. 4).

Volcanic forcing reconstructions before the satellite period are based on the analyses of acidity and sulfate measured in ice cores and catalogues of volcanic eruptions. Uncertainties arise because of the necessity to infer the spatiotemporal evolution of atmospheric optical depth changes from the limited and geographically biased indirect evidence. Large spikes of negative radiative forcing have been reconstructed (28) (Fig. 4). However, the bulk of the forcing for individual events is limited to a few years by the residence time of the injected particles in the lower stratosphere. The fundamentally different time scales governing volcanic versus anthropogenic forcing makes any direct comparison of rates difficult. On the annual time scale, maxima in volcanic radiative forcing and its rate of change are clearly larger than those from greenhouse gases. In an attempt to compare the forcings also on the decadal time scale, we spline the volcanic record with the same cutoff period of 40 years as the other records, thereby implicitly assuming that the spikes in volcanic forcing express themselves also in forcing and climate variations on decadal time scales. This assumption is justified by results from climate models (26). The inferred decadal-scale forcing is <1 W m^{-2}. Decadally smoothed volcanic forcing has been more negative since 1964 than during the previous 100 years. This is consistent with the finding that natural forcing has exerted a cooling influence on the global surface during recent decades (26).

The decadal-scale rates of change in forcing from explosive volcanism show a strong oscillatory behavior. However, the phases with positive decadal-scale rates of change in volcanic forcing last, for our data treatment, only up to 35 years and are preceded by negative growth rates. In contrast, the rate of change has been positive over the last 250 years for the greenhouse gas forcing. The largest average rate of increase in volcanic forcing over any 40-year period of the past millennium, as inferred from the spline, is 15 × 10^{-3} W m^{-2} yr^{-1}. The corresponding rate for the combined solar (27) and volcanic (28) forcing is another 5 × 10^{-3} W m^{-2} yr^{-1} higher. This suggests that the average rate of increase in anthropogenic forcing of the past 40 year may be larger than the rate of increase in natural forcing for any 40-year period of the past millennium.

In summary, the rate of change in anthropogenic greenhouse gas forcing is unique in the context of the past. The 20th century rise in anthropogenic forcing occurs faster than changes in the combined radiative forcing from CO_2, CH_4, and N_2O during the past 20,000 years. Decadal-scale rates of change in CO_2, CH_4, and N_2O and in their forcing are several times larger during the Industrial Era than the last 2,000 years. In addition, the multidecadal scale rate of change in anthropogenic forcing is also very high in the context of the known natural forcing variations of the past millennium. For recent years, the analysis by (7) shows that CO_2 emission from fossil and industrial sources, the primary driver of anthropogenic climate forcing, have been accelerating over the past few years compared with the 1990s.

Regarding climate change, the cause-effect chain from anthropogenic emissions to atmospheric concentrations to radiative forcing to climate change (13) implies that the ongoing anthropogenic climate change very likely proceeds with a high speed compared with naturally forced decadal-to-century scale global climate variations of the past millennia. Data and models show that the global warming of 4 to 7°C since the Last Glacial Maximum occurred at an average rate ≈10 times slower than the warming of the 20th century (5). An increase in global mean temperature of up to 6°C is projected for business-as-usual scenarios over this century (29). This is comparable in magnitude to the millennial-scale increase from the last ice age to the current interglacial, but is projected to occur within 100 years only. Our analysis of forcing supports the conclusion that human society and the socioeconomic and natural systems are confronted with global climate change progressing rapidly.

Material and Methods

Concentration and Radiative Forcing Data. Ice core and atmospheric greenhouse gas data for CO_2, CH_4, and N_2O are compiled from a variety of sources (see *SI Text*) to construct continuous records for the past 2,000 years (SI Fig. 5) and the past 22,000 years (Fig. 1 and SI Fig. 6).

Concentration and emission histories are converted into radiative forcing using simplified expressions (13, 30) as summarized in ref. 31 and updated by ref. 32 (SI Table 3 and *SI Text*). For comparison, the evolution of atmospheric sulfate aerosol loading and forcing since 1765 are estimated from statistics on past emissions (22) as corrobated by measurements of sulfate deposition on ice sheets and ice caps (e.g., ref. 33); total tropospheric sulfate aerosols forcing is assumed to be −1.1 W m^{-2} in year 2000, consistent with satellite-based estimates and indirect approaches (e.g., ref. 34). Histories of 13 gases controlled under the Montreal Protocol, including CFC-11 and CFC-12, of SF_6, and of five other halocarbons were compiled as described in ref. 31 and based on refs. 35 and 36. Radiative forcing from explosive volcanisms are available for the past millennium (14, 28). Changes in solar irradiance are reconstructed from satellite data for the past two decades, sun spot observations for past centuries, and from the radioisotope ^{10}Be (15, 27).

Spline Fitting Procedure. The ice core and atmospheric concentration records are fitted with a spline (18). The resulting continuous records are then used to calculate radiative forcing and rates of change in concentration and forcing (*SI Text*).

The records of the past 2,000 years are smoothed with a cut-off period of 40 years, guided by the width of the age distribution in the Law Dome cores of up to 20 years, to retain multidecadal variability. The atmospheric methane record shows a strong decrease in the growth rate over the recent decades. To include this decrease, a cutoff period of 10 years is selected for methane and for 1983–2004, the period with direct atmospheric CH_4 samples (This results in lower rates for recent years than a cutoff of 40 years). The CO_2, CH_4, and N_2O records of the past 22 ka are smoothed with different cutoff periods for different time periods to follow the variations in the ice core records, taking into consideration data resolution. For comparison, the GRIP CH_4 record, with a small width of the age distribution, has been splined separately to retain multidecadal to century-scale variations over the past 22 ka (Fig. 1c and SI Fig. 6).

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