

catalyst layer in any case, even for the low-temperature experiments (fig. S6B). The coking on Ru was presumably kinetically limited.

There are three minor drawbacks to the catalyst layer. First, it is expected to reduce the rate at which fuel can diffuse to the anode, thereby decreasing cell power density. Indeed, test results at 770°C typically showed lower power densities of ~ 0.6 W/cm² with the catalyst layer compared with initial values (prior to degradation) of 0.8 to ~ 1 W/cm² without the layer. Second, because the catalyst layer was electrically insulating in the present experiments, electrical current collection could be an issue. However, a slight modification of a typical interconnect design, in which the catalyst layer is present only in the interconnect gas-flow channels as shown schematically in fig. S7 (4), could be used to provide good current collection. Third, Ru is expensive, although less so than precious metal catalysts such as Pt and Rh. As discussed in the supplemental materials (4), this cost should not be prohibitive for reasonable Ru loadings.

The other factor required for stable cell operation was having at least 10% air in the fuel mixture. This represents a 2% oxygen addition to the fuel, and it is not clear what role this plays in preventing coking. One possibility is that the oxygen helps remove carbon on the catalyst. Although the addition of oxygen amounts to burning some of the fuel, the amount of air is too small to substantially reduce the efficiency or to substantially dilute the fuel with nitrogen.

The present results compare favorably with other recently reported methods for using heavy hydrocarbon fuels in SOFCs. SOFCs have recently been reported to operate successfully on N₂-diluted gasoline (18) and other heavy hydrocarbon fuels (20). However, the power densities were substantially lower (0.1 W/cm²) than in the present results. Prototype SOFC systems have recently been developed for vehicle auxiliary electrical power using external partial-oxidation reforming of gasoline to produce a hydrogen-rich fuel (6). However, partial-oxidation reforming has substantially lower efficiency than is possible with H₂O-CO₂ reforming (21). In cases where reforming is done within the stack but away from the cells (22) or in an external reformer (23), heat transfer is not as good and additional hardware is required. One disadvantage of direct internal reforming is that the endothermic reaction may be too rapid and may cause substantial SOFC or catalyst cooling near the fuel inlet (24). The present catalyst layer may have an advantage in this regard, because the catalyst material can be varied to suitably adjust the rate of the reforming reaction. The addition of oxygen to the fuel also helps mitigate this cooling because of the exothermic partial-oxidation reaction.

The present small-scale demonstrations show the feasibility of SOFCs fueled by hydrocarbons such as *iso*-octane, with substantial advantages including high efficiency, a relatively simple system design, and reduced operating temperature. However, more work needs to be done to prove that this approach is practical with real fuels such as gasoline, diesel, and aircraft fuels; recently developed techniques for reducing sulfur contamination to low levels (25) may be especially useful.

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Supporting Online Material

www.sciencemag.org/cgi/content/full/1109213/DC1
Materials and Methods
SOM Text
Figs. S1 to S7
References

28 December 2004; accepted 15 March 2005

Published online 31 March 2005;

10.1126/science.1109213

Include this information when citing this paper.

From Dimming to Brightening: Decadal Changes in Solar Radiation at Earth's Surface

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Variations in solar radiation incident at Earth's surface profoundly affect the human and terrestrial environment. A decline in solar radiation at land surfaces has become apparent in many observational records up to 1990, a phenomenon known as global dimming. Newly available surface observations from 1990 to the present, primarily from the Northern Hemisphere, show that the dimming did not persist into the 1990s. Instead, a widespread brightening has been observed since the late 1980s. This reversal is reconcilable with changes in cloudiness and atmospheric transmission and may substantially affect surface climate, the hydrological cycle, glaciers, and ecosystems.

Solar radiation at Earth's surface (also known as global radiation or insolation) is the primary energy source for life on our planet. Widespread measurements of this quantity began in the late 1950s. Trends in worldwide distributed observational records of solar radiation have been proposed in various studies (1–5). These studies report a general decrease of sunlight over land surfaces on the order of 6 to 9 W m⁻² from the beginning of the measurements in about 1960 until 1990, cor-

responding to a decline of 4% to 6% over 30 years. Such a decrease may profoundly influence surface temperature, evaporation, the hydrological cycle, and ecosystems, as noted in (6–10).

Thus far, no study has addressed the evolution of solar radiation from 1990 onward, because extensive observational data after 1990 were not easily accessible. The main source for data prior to 1990 in (1–5) was the Global Energy Balance Archive (GEBA) (11), which

Table 1. Changes in surface solar radiation over Europe. Three hundred sites were merged into 32 ISCCP (27) equal-area grid cells over Europe. Results were obtained by fitting linear models with station effects (2) to annual means of surface solar radiation within each cell for two specified periods (significant trends at the 5% level are in parentheses). The period 1950 to 1990, considered in earlier studies (1–5), predominantly shows decreases in surface solar radiation, whereas increases dominate in the period 1985 to 2000. Data source: GEBA/WRDC (11).

	1950–1990	1985–2000
Number of cells	32	32
Increase	8 (3)	26 (8)
Decrease	24 (13)	6 (0)

we have updated for the 1990s in the present work, with support from the World Radiation Data Centre (WRDC) in Saint Petersburg, Russia. We also used surface radiation measurements from the Baseline Surface Radiation Network (BSRN) of the World Climate Research Program (WCRP), available from 1992 onward (12). This global network measures surface radiative fluxes at the highest possible accuracy with well-calibrated state-of-the-art instrumentation at selected sites in the major climate zones. The data in both GEBA and BSRN underwent rigorous quality checks, as described in (11, 12), to assure high accuracy as well as homogeneity in the data, a prerequisite for regression analyses. Here, we evaluate the newly available surface observations to investigate changes in solar radiation in more recent years.

The most comprehensive data for the 1990s are available for the European area. Seven thousand yearly values measured at 300 stations from GEBA/WRDC were analyzed in 32 grid cells on an equal-area grid of ~2.5° resolution. The results in Table 1 were obtained by estimating linear models in each cell [including station effects, as described in (2)]. Out of 24 cells with a systematic decrease over the period 1950 to 1990 considered in the earlier studies (1–5), only 6 show a decrease over the period 1985 to 2000, none of them statistically significant. As can be inferred from Table 1, dimming of solar radiation fades after 1985 over Europe, and a

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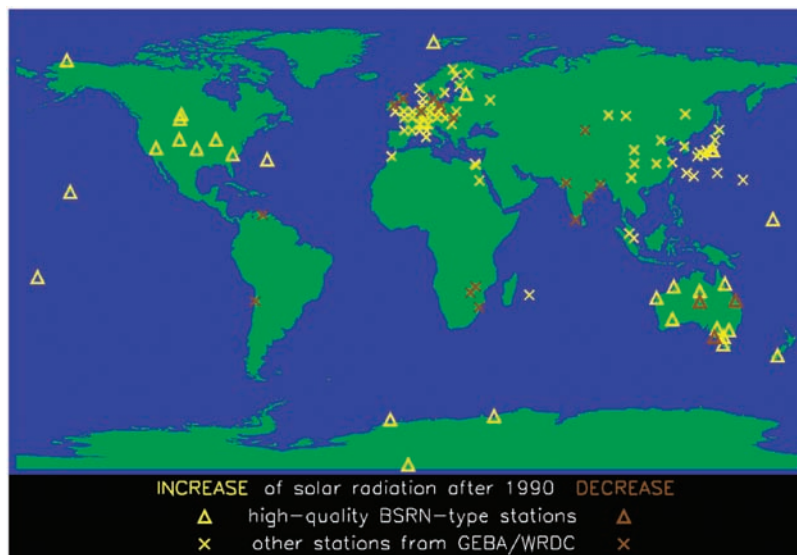


Fig. 1. Global distribution of surface observation sites used in this study. Sites measuring an increase in surface solar radiation after 1990 are marked in yellow; sites measuring a decrease are shown in brown. High-quality observation sites fulfilling the BSRN standards (12) are shown as triangles, other sites from the updated GEBA as crosses. Information from 300 sites over Europe and 45 sites over Japan are displayed as aggregated regional means. The majority of the sites show an increase in surface solar radiation after 1990.

reversal to brightening is found (13). On average, trends change their sign from negative to positive in 1985, as obtained from the minima of second-order linear models in 14 cells. Illustrations for the reversal from dimming to brightening at various sites in Central and Eastern Europe are found in figs. S1 to S3.

The transition from decreasing to increasing solar radiation is in line with a similar shift in transparency of the cloud-free atmosphere determined from pyrhelimeter measurements, which show a general tendency of decreasing atmospheric transmission up to the early 1980s and a gradual recovery thereafter (fig. S4). This may be related to a decrease of aerosol burden due to more effective clean-air regulations and the decline in the economy with the political transition in Eastern European countries in the late 1980s, as manifested, for example, in a lower local planetary albedo due to reduced aerosol loadings and related effects on clouds in these countries (14). Associated changes in atmospheric transmission, solar radiation, and cloudiness are documented in the long-term records from Tartu-Toravere in Estonia (figs. S3 and S4) (15).

In addition to these European-based observations, we found a similar reversal from dimming to brightening in multidecadal observational records around the world (Fig. 1 and figs. S5 to S11). These include the carefully calibrated and maintained sites of the Climate Monitoring and Diagnostics Laboratory (CMDL) (16) located in North America (Boulder, Colorado, and Barrow, Alaska), in the North and South Pacific (Mauna Loa, Hawaii, and Samoa), and in Antarctica (South Pole). The CMDL sites show a recent recov-

ery from their downward tendencies before the mid-1980s (Fig. 1 and fig. S5).

The available data in the former Soviet Union allow the analysis of solar radiation up to 1996 for the Moscow region (17) (fig. S6). A change from decrease to increase in solar radiation is found during the 1980s, in line with atmospheric transmission measurements in fig. S4. For Japan, the comprehensive data available for the 1990s also suggest a recovery from the prior dimming (Fig. 1 and fig. S7). A strong increase in insolation during the 1990s is apparent in the longest time series available from Japan (Tateno), which goes back to 1958 (fig. S8). This is in line with the increasing atmospheric transmission since the 1980s documented in a unique data set from 14 pyrhelimeter sites in Japan (fig. S4). The majority of the sites in China, available in GEBA since 1988, show an increase in insolation as well (Fig. 1 and fig. S9). This supports findings of a recent study based on 85 rural radiation sites in China, where the decline of solar radiation between the 1950s and 1980s levels off in the 1990s [see figure 6 in (18)]. A reversal from dimming to brightening during the 1980s is further found at sites in Singapore and Malaysia (Fig. 1 and fig. S10).

A high-quality radiation network was established in 1993 in Australia. Data available up to 2003 do not support a continued dimming, because a majority of the sites show an increase in solar radiation in recent years (Fig. 1 and fig. S11).

Indications of a significant continued dimming in the 1990s are largely restricted to data from India [based on limited data that passed the quality checks (fig. S12)], possibly related

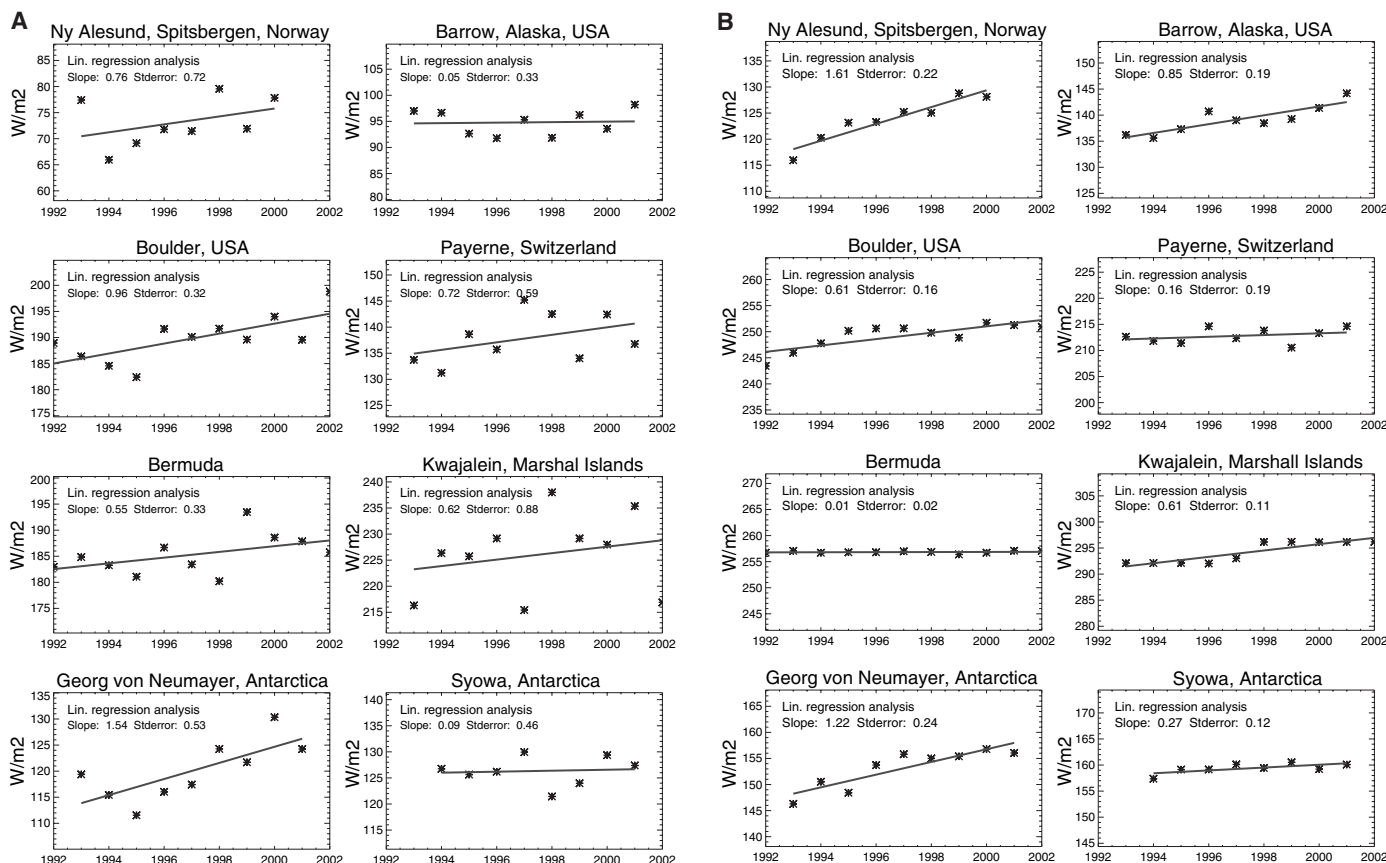


Fig. 2. Time series of annual mean surface solar radiation measured at worldwide distributed sites from BSRN. Shown are the eight longest records from BSRN covering the period 1992 to 2002 for (A) all-sky

conditions and (B) clear-sky conditions (24). Solar radiation increases at all sites under both all-sky and clear-sky conditions over this period. Units $W m^{-2}$.

to the ongoing prevalence of atmospheric brown clouds (ABCs) (19). In addition, on the African continent, surface solar radiation decline measured at two sites in Zimbabwe shows no tendency for recovery, whereas the dimming in Egypt levels off during the 1990s (Fig. 1 and figs. S13 and S14).

Data available from various other places do not reach the level of accuracy that would allow for time-series analysis, including sites in South America, Africa, and the United States (during the 1980s), as well as the Australian data prior to 1988; the latter were model-adjusted and thus artificially trend free.

This rather unsatisfactory situation with data quality led to the establishment of BSRN in 1992, where new quality standards for solar radiation measurements were introduced. From BSRN, we selected the sites with the longest available records, including the high-latitude sites Ny Alesund (Spitsbergen, Norway) and Barrow (Alaska, USA) in the Arctic as well as the Georg von Neumayer and Syowa stations in Antarctica, the mid-latitude sites Boulder (Colorado, USA) and Payerne (Switzerland), and the low-latitude sites Bermuda (West Atlantic) and Kwajalein (Tropical West Pacific). Time series of annual mean surface solar radiation at these sites are shown in Fig. 2A with their associated linear fits. It is noteworthy that

none of the sites shows a decline. Rather, six of the eight sites show a substantial increase. Similar tendencies are found at other BSRN sites with shorter time series (Fig. 1). Thus, the highest quality data available for the 1990s suggest a brightening rather than a dimming.

Overall, the information contained in the GEBA/WRDC, BSRN, and CMDL records provides no evidence for the continuation of widespread dimming into the 1990s. Instead, there are indications for an increase in surface insolation since the mid-1980s at many locations, mostly in the Northern Hemisphere but also in Australia and Antarctica. A similar reversal to brightening in the 1990s has been found on a global scale in a recent study that estimates surface solar radiation from satellite data (20). This indicates that the surface measurements may indeed pick up a large-scale signal. The changes in both satellite-derived and measured surface insolation data are also in line with changes in global cloudiness provided by the International Satellite Cloud Climatology Project (ISCCP) (21), which show an increase until the late 1980s and a decrease thereafter, on the order of 5% from the late 1980s to 2002. A recent reconstruction of planetary albedo based on the earthshine method (22), which also depends on ISCCP cloud data, reports a similar decrease during the

1990s. Over the period covered so far by BSRN (1992 to 2001), the decrease in earth reflectance corresponds to an increase of $6 W m^{-2}$ in absorbed solar radiation by the globe (22). The overall change observed at the BSRN sites, estimated as an average of the slopes at the sites in Fig. 2A, is $0.66 W m^{-2}$ per year ($6.6 W m^{-2}$ over the entire BSRN period). The dramatic increase in the planetary albedo estimated in (22) for 2002/2003 lies outside the period of available surface measurements and is controversial (23).

A further advantage of the BSRN data is their high temporal resolution (minute means), in contrast to conventional radiation data typically available in the form of monthly or daily means. The high-frequency measurements allow a stratification of the BSRN records into cloudy and clear-sky periods on the basis of an advanced clear-sky detection algorithm (24). The availability of extended records under both clear- and all-sky conditions for different climatic regimes provides a unique opportunity to study the transmission of solar radiation through the atmosphere. In Fig. 2B, time series of clear-sky insolation aggregated into annual means are shown with their associated linear fits for the eight BSRN sites, with slopes ranging from $+0.01$ to $+1.61 W m^{-2}$ per year. This suggests that the cloud-free atmosphere

might have become more transparent during the 1990s, in line with atmospheric transmission measurements in fig. S4. During the early 1990s, the increase in atmospheric transmission reflects the recovery from Pinatubo aerosol loadings. In addition, air-quality regulations and the decline of the Eastern European economy may have affected the large-scale aerosol concentration (25). The overall increase in the clear-sky fluxes, again estimated as an average over the slopes at the sites in Fig. 2B, is 0.68 W m^{-2} per year, comparable to the increase under all-sky conditions. The similar changes under clear- and all-sky conditions indicate that, besides clouds, changes in the transparency of the cloud-free atmosphere also contributed to the increase in insolation.

To summarize, our data suggest that the widespread decline of solar radiation widely reported for the period of about 1960 to 1990 did not continue in the following years. Rather, there are indications that the amount of sunlight at the surface has increased during the 1990s at most of the locations for which good records exist. This is found under all- and clear-sky conditions, indicating that processes in both cloud-free and cloudy atmospheres contributed to the brightening during the 1990s, possibly pointing to an interplay of direct and indirect aerosol effects.

The absence of dimming since the mid-1980s may profoundly affect surface climate.

Do Satellites Detect Trends in Surface Solar Radiation?

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Long-term variations in solar radiation at Earth's surface (S) can affect our climate, the hydrological cycle, plant photosynthesis, and solar power. Sustained decreases in S have been widely reported from about the year 1960 to 1990. Here we present an estimate of global temporal variations in S by using the longest available satellite record. We observed an overall increase in S from 1983 to 2001 at a rate of 0.16 watts per square meter (0.10%) per year; this change is a combination of a decrease until about 1990, followed by a sustained increase. The global-scale findings are consistent with recent independent satellite observations but differ in sign and magnitude from previously reported ground observations. Unlike ground stations, satellites can uniformly sample the entire globe.

The concept of "global dimming" (1–3), which refers to long-term measured decreases in the amount of solar radiation that reaches Earth's surface (S), has received prominent

attention because of concerns about its possible climatic and environmental implications. An early report on this topic based on surface observations made primarily in Europe (4) suggested that S declined by more than 10% from 1960 to 1990. On the basis of the analysis of a more comprehensive observational database, it was shown that over land, S decreased on the average by 0.23% (1) and 0.32% (2) per year from 1958 to 1992. The largest decrease was in parts of the former Soviet Union (5), where S decreased by about

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Supporting Online Material

www.sciencemag.org/cgi/content/full/308/5723/847/DC1
Figs. S1 to S14

26 July 2004; accepted 16 March 2005
10.1126/science.1103215

20% between 1960 and 1987. Independent indirect evidence for plausible decreases in S has been found in pan evaporation records (6, 7), which show that the rate of evaporation did not increase but rather decreased, in spite of global warming trends evident in records of surface temperatures. When the evaporation data were compared with the global dimming records, the respective tendencies matched, which suggests that these two processes might be linked. Two other studies (8, 9) found that S in the Swiss Alps increased between 1995 and 2003 after decreasing from 1981 to 1995 (8).

Speculations about possible causes of global dimming include cloud changes, increasing amounts of human-made aerosols, and reduced atmospheric transparency after explosive volcanic eruptions. (Data indicating global dimming could also be produced by instrument deficiencies.) Particles of soot and sulfates absorb and reflect sunlight and facilitate the formation of larger and longer-lasting clouds. The Indian Ocean Experiment (10) has clearly documented the large, short-term reduction in solar radiation reaching the surface caused by absorbing aerosols, particularly black carbon and dust. Regionally, the seasonally averaged reduction in the Indian Ocean can reach 10 to 30 W m^{-2} . However, there is some evidence that the reported longer-term

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