REVIEW

Key Points:
- Sunshine records suitability to detect atmospheric aerosol is reviewed
- Despite short comings of the data, aerosol effect on sunshine is evident
- Possible directions for future research are highlighted

Correspondence to:
A. Sanchez-Romero,
alejandro.sanchez@udg.edu

The signal of aerosol-induced changes in sunshine duration records: A review of the evidence

A. Sanchez-Romero[1], A. Sanchez-Lorenzo[1], J. Calbó[1], J. A. González[1], and C. Azorin-Molina[2]

[1]Department of Physics, University of Girona, Girona, Spain, [2]Instituto Pirenaico de Ecología, Consejo Superior de Investigaciones Científicas (IPE-CSIC), Zaragoza, Spain

Abstract Aerosols play a significant yet complex and central role in the Earth's radiation budget, and knowledge of long-term changes in the atmospheric turbidity induced by aerosols is therefore fundamental for a better understanding of climate change. However, there is little available information on changes in aerosol concentration in the atmosphere, especially prior to the 1980s. The present paper reviews publications reporting the suitability of sunshine duration records with regard to detecting changes in atmospheric aerosols. Some of the studies reviewed propose methods for estimating aerosol-related magnitudes, such as turbidity, from sunshine deficit at approximately sunrise and sunset, when the impact of aerosols on the solar beam is more easily observed. In addition, there is abundant evidence that one cause of the decadal changes observed in sunshine duration records involves variations in atmospheric aerosol loading. Possible directions for future research are also suggested: in particular, detailed studies of the burn (not only its length but also its width) registered by means of Campbell-Stokes sunshine recorders may provide a way of creating time series of atmospheric aerosol loading metrics dating back to over 120 years from the present.

1. Introduction

Solar radiation reaching the Earth's surface, also known as surface downward shortwave radiation, plays a key role in the global energy balance and therefore in modulating climate [see, for example, *Sellers*, 1965]. Apart from the atmospheric gases (including greenhouse gases (GHG)) affecting the radiative transfer of shortwave and longwave radiation through the atmosphere, clouds and aerosols are the main factors modulating the planetary energy balance [e.g., *Iqbal*, 1983; *Twomey*, 1974]. Thus, clouds and aerosols significantly contribute to the global albedo and, at regional scales, may produce warming or cooling effects depending on their characteristics [e.g., *Ramanathan et al.*, 1989; *Hartmann et al.*, 1992].

An atmospheric aerosol involves suspension of solid and/or liquid particles in the air [*D'Almeida et al.*, 1991]. The term "atmospheric aerosol" encompasses a wide range of particles (aerosols) with different compositions, sizes, shapes, and optical properties. Both natural and human processes contribute to the presence of aerosols in the atmosphere. The concentration of aerosols alters the intensity of solar radiation scattered back to space, absorbed in the atmosphere, and reaching the Earth's surface [e.g., *McCormick and Ludwig*, 1967; *Charlson et al.*, 1992; *Moosmüller et al.*, 2009]. The optical effect resulting from the absorption and scattering by aerosols, along with water in the liquid/ice phases, is known as atmospheric turbidity, which is an important atmospheric property in climate and pollution studies, as well as in aspects relating to solar energy [*Power*, 2003a]. Atmospheric turbidity is determined by measuring spectral (wavelength-specific) or broadband solar radiation. Several atmospheric turbidity indices have been used for almost one century to quantify the influence of atmospheric aerosol content on the radiation received at the Earth's surface [for an overview, see *Eltbaakh et al.*, 2012].

On a global average basis, aerosol radiative forcing is negative (i.e., it has a cooling influence) [*Solomon et al.*, 2007]. However, because of the spatial and temporal nonuniformity of the aerosol radiative forcing and due to the differences in its shortwave and longwave forcings, the net effect of aerosols on Earth's climate does not involve a global offsetting of the warming impact of anthropogenic GHG emissions [e.g., *Charlson et al.*, 1992; *Power*, 2003a]. More specifically, aerosols affect Earth's energy budget in several ways [e.g., *Ramanathan et al.*, 2001; *Rosenfeld et al.*, 2008], which are referred to as direct, semidirect, and indirect aerosol effects. A detailed overview of these aerosol radiative mechanisms is provided by *Haywood and Boucher* [2000].

One of the major pieces of evidences supporting the crucial role played by the direct and indirect effects of aerosols upon the climate system is provided by the dimming and brightening phenomenon [e.g., *Stanhill and Cohen*, 2001; *Stanhill*, 2005; *Wild*, 2009, 2012]. Both terms refer to a widespread decrease (dimming) in downward shortwave radiation from the 1950s to the 1980s [*Stanhill and Cohen*, 2001], while a reversal of this trend (brightening) has been seen since the late 1980s, particularly in the developed countries [*Wild et al.*, 2005; *Wild*, 2012]. Similar trends have also been observed when only cloudless conditions are considered [e.g., *Qian et al.*, 2007; *Ruckstuhl et al.*, 2008]. The causes of the dimming/brightening are very complex, although changes in atmospheric transmissivity since the mid-20th century resulting from variations in anthropogenic aerosol emissions are considered as the main factor causing the observed trends in solar radiation [*Stanhill and Cohen*, 2001; *Wild*, 2009, 2012]. In particular, an increase in these emissions was observed from the 1950s to the 1980s, i.e., during the period when the global dimming is detected. A subsequent decrease in anthropogenic aerosol emissions from the 1980s to the 2000s has been reported, resulting from actions aimed at regulating air pollution in the developed countries, which coincide with the period of brightening [e.g., *Stern*, 2006; *Streets et al.*, 2009; *Wild*, 2009; *Folini and Wild*, 2011; *Smith et al.*, 2011].

Nevertheless, uncertainties remain regarding the causes of the global dimming/brightening phenomenon [e.g., *Wild*, 2012]. In addition, there is little availability of long-term measurements of downward shortwave radiation prior to the mid-20th century when widespread measurements were initiated within the framework of the International Geophysical Year (1957–1958). Measurements of atmospheric turbidity are even scarcer, and instrumentation for observations of aerosols has been developed more recently. All these factors limit the spatial and temporal representativeness of the observed trends. It would therefore be useful to find a proxy for these magnitudes with greater spatial and temporal coverage.

Since long time series of sunshine duration (SD) measurements exist, they have a remarkable historical value. According to the *World Meteorological Organization* (*WMO*) [2008], the SD for a given period is currently defined as the sum of those subperiods for which the direct solar irradiance exceeds $120 \, \mathrm{W m^{-2}}$. For climatological purposes, the units used are "hours per day," as well as percentage quantities, such as "relative daily sunshine duration," where SD is divided by the maximum possible SD (i.e., as if sky was clear all the time). SD is affected by atmospheric conditions, and consequently, it could serve as a proxy for atmospheric turbidity. Another proxy of atmospheric turbidity that has often been employed is visibility [*Wang et al.*, 2009; *Zheng et al.*, 2011; *Van Beelen and Van Delden*, 2012]. The problem with visibility is, apart from being a magnitude even more subjective than SD, that few available long-term series exist.

One of the instruments used to measure SD is the Campbell–Stokes sunshine recorder (CSSR). It was invented in the late nineteenth century to provide a measurement of the duration of bright sunlight by making a burn mark on a piece of specially treated cardboard. The measurement of the length of the burn for a given card gives daily SD and also provides additional embedded information on cloudiness and solar irradiance [*Stanhill*, 2003; *Wood and Harrison*, 2011]. Regarding the latter possibility, many authors relate SD during a period of time with direct or global (which is the sum of the solar direct and diffuse contributions) irradiance, with the use of Ångström–Prescott type formulas [e.g., *Martínez-Lozano et al.*, 1984; *Stanhill*, 1998b; *Power*, 2001; *Suehrcke*, 2000; *Bakirci*, 2009], which were first proposed by *Ångström* [1924] and further modified by *Prescott* [1940]. Moreover, several studies have analyzed the spatial and temporal behavior of SD in different regions during the last few decades [e.g., *Weber*, 1990; *Liu et al.*, 2002; *Brázdil et al.*, 1994; *Pallé and Butler*, 2001; *Stanhill and Cohen*, 2005, 2008; *Sanchez-Lorenzo et al.*, 2007, 2008; *Raichijk*, 2012; *Sanchez-Lorenzo and Wild*, 2012; *Wang et al.*, 2012a].

The possibility that SD records can be affected by the changes in atmospheric turbidity and thus by the aerosol loading was proposed by J. F. Campbell, the inventor of the CSSR. Specifically, he claimed [*Campbell*, 1857, p. 1]: "The bowl, at the Board of Health—*downtown London*, which is always in the smoke, was not nearly so much marked as the one at Campden Hill—*high ground in west London*. One further from London would probably have been marked to a much greater extent and for much longer periods…" Later, other early studies found more evidence that air pollution can affect the measured SD. For example, *Whipple* [1878] compared SD data from two cities to the east and west of London and found, by analyzing the reduction of the length of the burn, reductions in air transparency depending on wind direction. A large number of studies show a relationship between SD and smog in London during the last century [for an overview, see *Wheeler and Mayes*, 1997]. Equally, *Maurer and Dorno* [1914] already discussed how the eruption of Katmai (Alaska) in
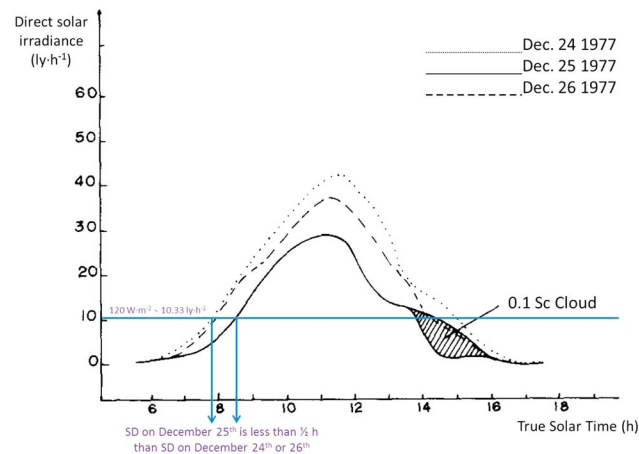
Figure 1. The large quantity of aerosol, emitted during the night of 24 December 1977 in Mexico City, produced a reduction of direct solar irradiance in the morning of the next day, and in consequence, the paper strip of the CSSR started to burn later than the other days. Adapted from *Galindo and Chavez* [1978]. © Geofísica Internacional. Used with permission.

1912 caused anomalous records of SD in various European observatories during 5 months following the eruption. *Galindo and Chavez* [1978] reported an increase in the aerosol loading on the night of 24 December in Mexico City, caused by the burning of thousands of tires, causing a reduction by 1 h in SD the next day. Specifically, on 24 and 26 December, the cardboard started to burn at 0650; on 25 December, it started at 0750 (Figure 1).

R. Jaenicke and L. Helmes developed a series of pioneering studies presenting a method to determine atmospheric turbidity from SD records [*Jaenicke and Kasten*, 1978; *Helmes and Jaenicke*, 1984, 1985, 1986]. The same method was applied some years later [*Wu et al.*, 1990; *Balling and Idso*, 1991] but, to our knowledge, has not been used since then. However, the subject has recently aroused interest, and several studies have shown the potential of SD records for detecting changes in atmospheric turbidity, especially in clear-sky conditions and mainly during sunrises and sunsets [e.g., *Horseman et al.*, 2008; *Sanchez-Lorenzo et al.*, 2009, *Sanchez-Lorenzo et al.*, 2013; *Xia*, 2010; *Wang et al.*, 2012a].

This article is based upon a number of studies that consider SD to be a good proxy for turbidity, as well as aerosol content in the atmosphere. Section 2 provides a brief description of the different instruments and methods for measuring SD, with particular focus on the CSSR. In section 3, we review a number of works that show the effect of aerosols on the measurement of SD and other studies that point toward evidences of aerosol loading in SD data. Finally, in section 4, we comment upon whether or not evidence of an aerosol signal from SD measurements is real and propose further research in this sense.
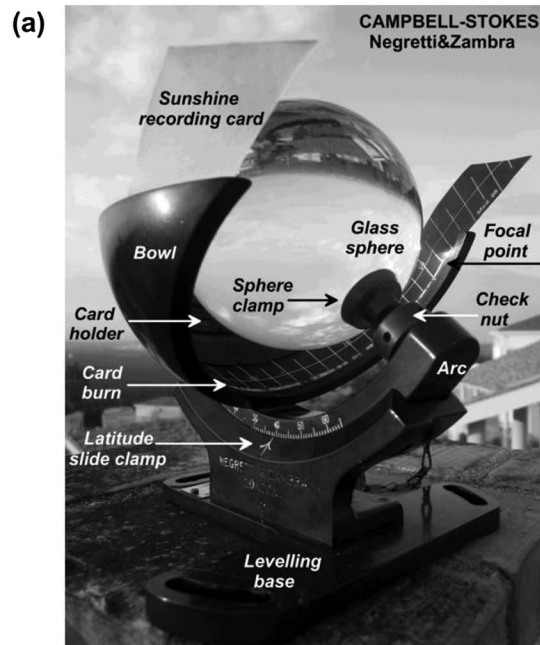
2. Sunshine Duration Measurements

In section 2.1, we present a brief summary of the main devices designed to measure SD, focusing on instruments developed since the late nineteenth century and the automation of SD measurements over the last few decades. The major problems when measuring burn lengths on CSSR sunshine cards are presented in section 2.2.

2.1. From the Pioneering Designs to the Current Methods

As mentioned by *Stanhill* [2003], perhaps the first sketch of a design for measuring SD can be attributed to Athanasius Kircher, [*Kircher*, 1646, p. 692; reproduced by *Stanhill*, 2003]. Kircher's design, which was never manufactured, showed a device with which the Sun's rays are focused by a glass sphere to ignite a bowl that is in the shape of a wooden chalice. Almost 200 years later, Thomas Brown Jordan (1807–1890) designed and manufactured the first known sunshine recorder, which was presented in 1838. The device was based on the effect of sunlight on a sensitive photographic paper controlled by a clockwork mechanism, but no systematic observations were performed due to the high cost of the instrument [*Jordan and Gaster*, 1886; *Maring*, 1897].

Subsequently, in 1853, John Francis Campbell (1821–1885) constructed a new instrument to measure the duration of the sunlight. The original design by Campbell consisted of a glass sphere filled with water set into a wooden bowl that was charred by the Sun's rays focused by the sphere. Four years later, in 1857, Campbell replaced the water lens by a compact spherical glass lens [*Campbell*, 1857]. In 1879, George Gabriel Stokes (1819–1903), a professor at the University of Cambridge, suggested changes in Campbell's original design, such as substituting the bowl with a metallic spherical segment with grooves to hold the cardboard cards
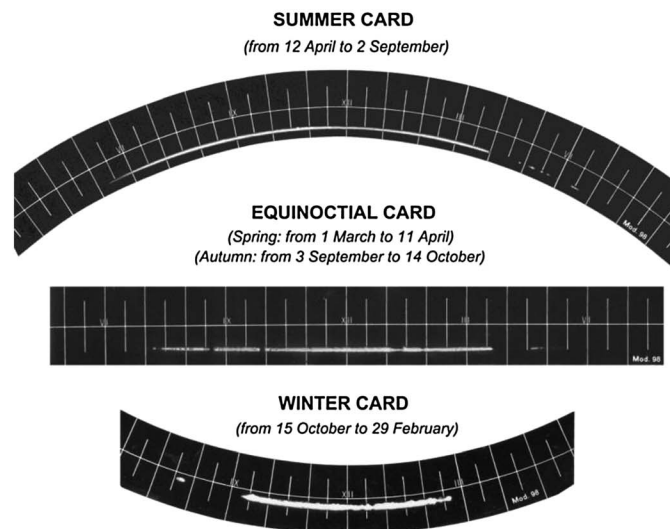
Figure 2. (a) Details of the different parts of a CSSR and (b) an example of the three types of cards used during summer, winter, and equinoctial seasons, respectively.

used to make daily measurements. This revised instrument was adopted at the Meteorological Office, and in the 1880s, more than 40 meteorological stations across the British Isles were reporting measurements of SD [*Scott*, 1885], most of them using Campbell's recorder with the modifications made by Stokes. For further details on the history of the CSSR, we refer to *Sanchez-Lorenzo et al.* [2013]. Figure 2a displays the main parts of a current CSSR, which involve a sphere (around 10 cm in diameter) made of high-quality, uniform, transparent glass and a rounded metal plate placed behind the sphere. The glass sphere is designed to focus the Sun's rays onto a piece of recording paper. The metallic spherical part has three overlapping sets of grooves of different length to hold the recording cards for the winter, summer, and spring/autumn periods (Figure 2b). The recording card should be replaced daily after sunset. Hourly and half hourly divisions are marked across it, enabling measurement of the times of sunshine (resolution of 0.1 h). For further details on the instrument and instructions for obtaining uniform results, see *Middleton* [1969] and *WMO* [2008].

In parallel to the development of the CSSR, during the mid-1880s, James B. Jordan and Frederic Gaster improved the instrument developed by Jordan's father in the 1830s. They introduced the new photographic sunshine recorder in 1885 and subsequently a new version of the device in 1888 [*Jordan*, 1888]. This latter instrument consisted of two hollow semicylinders used to contain the morning and the afternoon charts, respectively. A hole to let the beam of sunlight enter is made in the center of the side of each semicylinder, which leaves a mark on a strip of paper sensitized with ferrocyanide. A version of the recorder, slightly modified by C. F. Marvin in 1888, was introduced into routine SD measurements at some meteorological stations run by the United States Weather Bureau [*Maring*, 1897]. The Jordan photographic recorders were gradually replaced at the beginning of the twentieth century by the thermometric sunshine recorders designed by D. T. Maring and C. F. Marvin [*Brooks and Brooks*, 1947; *Stanhill and Cohen*, 2005]. The Maring–Marvin sunshine recorders were once again replaced in 1953 by new instruments based on a photoelectric switch, designed by N. B. Foster and L. W. Foskett [*Foster and Foskett*, 1953; *Michalsky*, 1992]. These in turn were removed from the meteorological stations in the late 1980s and early 1990s, which meant the end of the SD series in the United States.

During the last few years, various automated instruments and other methods for obtaining SD have been developed, which are summarized in *WMO* [2008]. First, we should mention the pyrheliometric and pyranometric methods [e.g., *Hinssen and Knap*, 2007; *Hinssen*, 2006; *Massen*, 2011]. The former are based on direct irradiance measurements, while the latter use global irradiance measurements. Another way of determining SD is by means of automatic instruments specifically designed to this end, which have become commercially available. These instruments are simple radiometers that detect direct solar radiation and count the time interval in which the irradiance exceeds a certain threshold. Progressively, many weather stations have changed traditional manual instruments (such as the CSSR and the Jordan photographic recorders) to these kinds of automatic sensors.

Kerr and Tabony [2004] compared SD from CSSR with automatic sensors and found that CSSR records tend to overestimate SD because the burn on the card spreads, especially when cloud cover is broken and the Sun is high in the sky. A change in the way to measure SD could therefore affect the homogeneity of the series and give rise to errors when evaluating trends [*Brázdil et al.*, 1994; *Stanhill and Cohen*, 2008]. Indeed, several studies assess the homogeneity of the SD series, such as research conducted in the Czech Republic [*Brázdil et al.*, 1994], Taiwan [*Liu et al.*, 2002], United Kingdom [*Kerr and Tabony*, 2004], Iberian Peninsula [*Guijarro*, 2007; *Sanchez-Lorenzo et al.*, 2007], Japan [*Katsuyama*, 1987; *Stanhill and Cohen*, 2008], China [*Xia*, 2010], and Switzerland [*Sanchez-Lorenzo and Wild*, 2012].

These new models of sunshine recorders do not require daily attention by an observer, data reduction (i.e., the process of filling and storing the SD data) is faster and more accurate, and routine and absolute calibration is possible [*Stanhill*, 2003]. All this appears to support the idea that automatic sensors will gradually replace the traditional instruments, but due to the additional information that can be extracted from burnt cards and their possible contribution to understanding climate change, there is a consensus with regard to preserving such instruments at long-established, well-maintained, and freely exposed meteorological stations [*Stanhill*, 2003; *Wood and Harrison*, 2011; *Sanchez-Lorenzo et al.*, 2013].

2.2. Variability of the Burning Thresholds

Differences between one type or another of SD measurements might be attributed to their particular characteristics and limitations. The two major problems with CSSR when comparing their measurements with other methods or instruments lie in the variability of the level of direct irradiance, which produces a burn and the overburning of the card in conditions of intermittent high irradiance [*Stanhill*, 2003]. These difficulties can be added to the obvious element of subjectivity in measuring the burn length on the sunshine cards [*Guijarro*, 2007].

The problem of overburning is very difficult to evaluate, as one small burst of high direct irradiance causes a burn lasting far longer than the few seconds of its actual duration, and standard methods have therefore been proposed to take into account this fact when evaluating the burn lengths [*WMO*, 2008]. Despite these rules, during events of very broken cloudiness, a measurement of SD by means of CSSR can be significantly overestimated [*Painter*, 1981; *Kerr and Tabony*, 2004].

Regarding the first problem, the direct irradiance value that produces burning ("burning threshold") is usually considered constant, i.e., $120 \, W/m^2$ [*Gueymard*, 1993; *WMO*, 2008]. Nevertheless, *Jaenicke and Kasten* [1978]

showed a large variety of burning thresholds for different CSSR. Similarly, *Helmes and Jaenicke* [1984] described the effects of using different types of recording cards. *Brázdil et al.* [1994] stressed the importance of using the same type of card in order to compare different SD series.

In order to homogenize the worldwide network, a specific design of the CSSR was recommended as the reference [*WMO*, 1962]. This however did not overcome all the problems. For example, *Bider* [1958], *Baumgartner* [1979], and *Painter* [1981] had observed that the burning threshold is on average higher in the early morning than in the late evening, thus producing notable losses of records, hypothetically attributed to dew or other water deposits on the glass sphere, since more energy is required to burn a trace in the card when conditions are cold and damp than when these are warm and dry. Equally, the absorption of water by the card can also affect the burning threshold. *Painter* [1981] also found that the burning threshold showed a marked seasonal effect; i.e., the burning threshold is higher in winter than in summer as a result of generally lower temperatures and higher relative humidity. Therefore, if global or direct radiation are to be estimated from SD records with the use of Ångström–Prescott type expressions, at the very least, we need to become aware of the limitations of the CSSR, in particular the burning threshold issue, and, where possible, we must address and correct them [*Roldán et al.*, 2005]. This issue should also be taken into account on considering the possibility of using SD records as a proxy for aerosol loading.

3. Evidence of Aerosol/Turbidity Effects on Sunshine Duration Measurements

In this section, we have reviewed empirical investigations dealing with the direct effects of aerosol on SD. These studies suggest methods for estimating atmospheric turbidity (section 3.1). In addition, we have also considered studies relating SD with meteorological and radiometric variables (clouds, global, direct and diffuse irradiance, wind speed, and relative humidity), where aerosols are suggested as an intervening factor (section 3.2).

3.1. Assessing Turbidity From CSSR Cards

A few articles focus on estimating the effect of aerosols in SD. Jaenicke, Kasten, and Helmes were the first authors to show interest in this sense. They proposed a method for quantifying atmospheric turbidity by means of data obtained from the readings of CSSR [*Jaenicke and Kasten*, 1978; *Helmes and Jaenicke*, 1984]. They based their arguments on the assumption that in the case of cloudless sunrises and sunsets, solar radiation is weakened because of the longer path it follows through the atmosphere. This weakening may suggest that the threshold value is not reached, and therefore, no trace is burnt on the card even if the Sun is visible. Their method is based on the Linke factor, T_L, which represents the number of clean and dry atmospheres that would be necessary to produce the same attenuation of the extraterrestrial radiation produced by the real atmosphere. T_L can be derived from the measured direct normal irradiance over the whole solar spectrum I [*Linke and Boda*, 1922]:

$$T_L = \frac{1}{m_A \delta_R} \ln\left(\frac{I_0}{I}\right) \tag{1}$$

where I_0 is the extraterrestrial solar irradiance, δ_R is the optical thickness of the clean dry atmosphere, and m_A is the relative optical air mass. Then, from a CSSR cardboard, T_L can be approximated by the linear function (valid for solar elevation angles above 5°) [*Jaenicke and Kasten*, 1978]:

$$T_L = (0.154\alpha + 1.05) \ln\left(\frac{I_0}{I_{CS}}\right) \tag{2}$$

where α is the solar elevation angle, calculated for the beginning or end of the burnt trace, and I_{CS} is the threshold value of the recorder. Equation (2) enables the computation of the turbidity factor T_L from the solar elevation α when burn starts, measured by any given SD recorder with known threshold irradiance I_{CS}. Experiments performed by *Jaenicke and Kasten* [1978] and *Helmes and Jaenicke* [1984] showed that equation (2) can be successfully used for SD data in cases of cloudless sunrises and sunsets (Figure 3). Nevertheless, a few sites provide information on cloudiness during sunrise and sunset, and therefore, only a few SD data can be used if cloudless conditions are to be guaranteed. Consequently, Helmes and Jaenicke developed another method for estimating atmospheric turbidity without requiring the condition of a cloudless sky [*Helmes and Jaenicke*, 1985, 1986]:
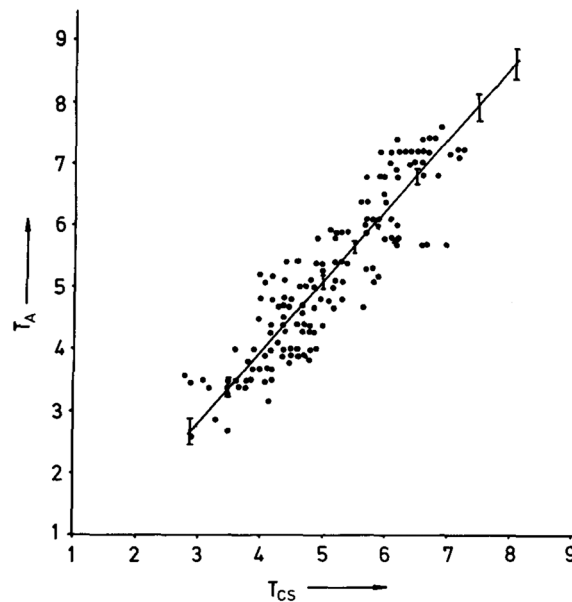
Figure 3. Turbidity determined with a pyrheliometer T_A as a function of turbidity computed with the burned trace method by *Helmes and Jaenicke* [1984], where T_{CS} is calculated using the individual threshold values I_{CS} for each type of the card determined by comparison with the pyrheliometer. Similar results are obtained using a fixed value of $I_{CS} = 20\,\text{mW cm}^{-2}$ for all cards. From *Helmes and Jaenicke* [1984]. © American Meteorological Society. Used with permission.

$$TCC + SD + T = 100\% \qquad (3)$$

where total cloud cover (TCC) was defined as the mean daily cloud cover in percentage of the sky area, SD as the sunshine duration in percentage of the astronomically possible, and T as the mean daily turbidity in percentage of the equivalent SD per astronomically possible SD [see *Helmes and Jaenicke*, 1985, 1986]. They found that the overall sum usually exceeded 100%, even if only TCC and SD were considered. They attributed this to an overestimation of cloud cover by most observers [e.g., *Van der Stok*, 1913; *Karl and Steuer*, 1990; *Jones and Henderson-Sellers*, 1991]. In order to overcome this issue, *Helmes and Jaenicke* [1985, 1986] proposed the following modification:

$$TCC + SD + T\frac{100 - TCC}{100} = f(TCC) \qquad (4)$$

where atmospheric turbidity is weighted by a factor that depends on TCC, in such a way that it ranges from one for cloudless days to zero for totally overcast days. The summation of the left-hand side terms is approximated by a linear function $f(TCC)$, which is empirically derived.

Thus, availing of daily data of TCC and SD, a value for daily atmospheric turbidity can be obtained even for cloudy days, with similar results as when only cloudless days are used. Although these methods are simple and give us an idea of the atmospheric turbidity, they pose the major problem that the turbidity index is affected not only by aerosols but also by water vapor.

The development of new, more precise instruments (pyrheliometers, narrowband photometers, etc.,), more remote sensing data, and the evolution of other methods have all caused discontinuity in the studies initiated by Jaenicke, Helmes, and their colleagues. Nevertheless, their methods have the advantage of being based on data which have been available for over 100 years; this justifies these methods for estimating atmospheric turbidity from SD in several studies. For instance, *Wu et al*. [1990] and *Balling and Idso* [1991], who were interested in volcanic material in the stratosphere, represented the variation of atmospheric turbidity obtained from sunshine records for Sonnblick (Austria) during the twentieth century. Using this station at high altitudes and far from cities, where boundary layer aerosols are unlikely to dominate the record, they found a clear decrease in stratospheric sulfate aerosols from the 1900s to the mid-1980s.

Horseman et al. [2008] assumed that high, thin cirrus clouds, and aerosols, only reduce SD at low solar elevations when incident direct solar radiation would not be much higher than $120\,\text{Wm}^{-2}$. Therefore, *Horseman et al*. [2008] described a method for extracting SD for cloudless periods at sunrise and sunset in order to track changes in pollution, using the cards of a CSSR from Lancashire (UK) during the 1976–2006 period. Unlike the method proposed by *Jaenicke and Kasten* [1978], this method is not a quantitative estimate of atmospheric turbidity; it does however provides a useful time series that can be interpreted in terms of changes of atmospheric opacity/pollution on daily basis. The method was proposed to test whether bright sunshine records at progressively lower solar elevation angles, corresponding to progressively longer path lengths, indicate progressively lower amounts of sunshine. This involves splitting the daily record of low-elevation SD (solar elevation between 2 and 8°) into a series of time subdivisions associated with increasing the path lengths of sunlight through the atmosphere and measuring an "efficiency of burn" in each of these subdivisions (three for winter and two for summer). Moreover, *Horseman et al*. [2008] divided the results into winter and summer periods, because the origins of particulate air pollution could differ: more pollution
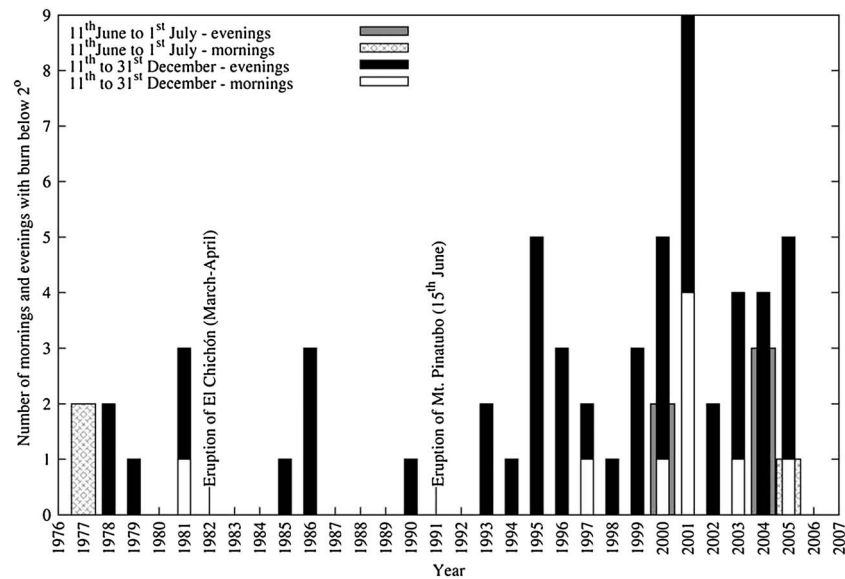
Figure 4. Counts of mornings and evenings within the analyzed solstice periods when burn(s) below 2° elevation was/were observed in each year. The totals are grouped into summer and winter. Note that after the early 1990s, there was an increase in the number of mornings and evenings per year when bright sunshine was recorded at sub-2° elevations. Note also that most of the increase was contributed by the winter period. From *Horseman et al.* [2008]. © Elsevier. Used with permission.

caused by space-heating-related emissions and more generated by photochemical processes occur during winter and summer, respectively. The results of the efficiency of burn at these pollution-sensitive angles showed an increase from the late 1980s to the early 2000s, which is in line with the widespread brightening found by *Wild et al.* [2005]. This brightening was more significant during winter than summer, which they related to a greater decrease in space-heating-related emissions than in photochemical pollution. Another interesting result involved the asymmetry of the daily burns between sunrise and sunset, which can be partly explained by the blocking of the morning horizon by topographic elevations, but may also be due in part to natural weather phenomena such as morning mist [*Bider*, 1958; *Painter*, 1981]. The current installation protocol for the sunshine recorders anticipates that no significant burns will be recorded below an elevation of 3°, but the results of *Horseman et al.* [2008] suggested that, although this assumption may have been true for most of the last century, there is now an increase in the number of the days when burns occur below a 2° elevation, associated with a decrease in pollution (Figure 4).

3.2. Aerosol Effects on SD Variability and Trends

In the last 2 decades, many papers have addressed the variability in SD, the trends thereof, and the possible causes of these. These studies used long-term SD series obtained from different sites throughout the world as summarized by *Sanchez-Lorenzo et al.* [2007]. As with the long-term measurements of downward shortwave radiation, most of the above-mentioned studies found a widespread decrease in SD between the 1950s and 1980s, i.e., the "global dimming" period. Equally, most of these studies also showed a partial recovery of SD since the 1980s, i.e., the "brightening" period. The impacts of the El Chichón (April 1982) and Pinatubo (June 1991) volcanic eruptions are also evident in some of the SD series, which present clear minima during the periods of 1982–1983 and 1992–1993, hypothetically due to the impact of the big increase in sulfate aerosols in the lower stratosphere, which reduces incoming solar radiation [e.g., *Sanchez-Lorenzo et al.*, 2009; *Sanroma et al.*, 2010; *Sanchez-Lorenzo and Wild*, 2012]. Despite the fact that cloudiness is the most important factor affecting SD, some research detects the aerosol signal by combining simultaneous time series of cloudiness and SD. Other studies claim that the effect of aerosols can also be detected by comparing solar radiation series with SD series. These studies therefore point to a detectable direct effect of aerosols on SD [*Stanhill and Cohen*, 2001; *Stanhill*, 2005; *Wild*, 2009]. Table 1 shows an overview of these papers and also gives a rough idea of the role the authors attributed to atmospheric aerosols. The reviewed publications summarized in Table 1 are detailed below.

Table 1. Summary of Works That Find Discrepancies Between Trends of SD and Trends of Cloud Clover and/or Surface Solar Radiation[a]

Author	Site	Stations	Period	Time Basis	SD Instrument[b]	Discrepancy[c]
Morawska-Horawska, 1985	Cracow (Poland)	1	1861–1980	Annual	CSSR	1950s–1980s
Kaiser, 2001	China	200	1954–1998	Seasonal and Annual	CSSR	1950s–1990s
Zheng et al., 2008	Southwest China	184	1961–2005	Annual	Jordan	1960s–2000s
Sanchez-Lorenzo et al., 2009	Iberian Peninsula	69	1961–2004	Seasonal and Annual	CSSR	1960s–1980s
Jaswal, 2009	India	40	1970–2006	Seasonal and Annual	CSSR	1970s–2000s
Sanchez-Lorenzo and Wild, 2012	Switzerland	17	1930–2010	Seasonal and Annual	CSSR	1980s–2000s
Wang et al., 2013	China	84	1955–2011	Seasonal and Annual	CSSR	1950s–2000s
Zheng et al., 2011	Southwest China	184	1961–2005	Annual	Jordan	1960s–2000s
Kaiser and Qian, 2002	China	200	1954–1998	Annual	Jordan	1950s–1990s
Changnon, 1981	Midwestern EE.UU (United States)	36	1901–1977	Seasonal and Annual	CSSR	1960s–1970s
Kitsara et al., 2013	Athens (Greece)	1	1951–2001	Seasonal and Annual	CSSR	1980s–2000s
Li et al., 2011	South China	147	1961–2005	Annual	CSSR	1960s–1990s
Xia, 2010	China	618	1955–2005	Seasonal and Annual	Jordan and CSSR	1950s–1980s
You et al., 2010	Tibetan Plateau	71	1961–2005	Seasonal and Annual	?	1980s–2000s
Brunetti et al., 2009	Greater Alpine Region	242	1886–2005	Seasonal and Annual	CSSR	1900s–1950s
Plantico et al., 1990	EE.UU	94	1900–1987	Seasonal and Annual	Jordan, M-M, and Foster	1900s–1950s
Liu et al., 2002	Taiwan (China)	25	1898–1999	Seasonal and Annual	CSSR, Jordan, and Eko	1960s–1990s
Yang et al., 2012	Tibetan Plateau	78	1984–2006	Seasonal and Annual	?	1980s–2000s
Power, 2003b	Germany	13	1977–2000	Annual	CSSR	1970s–1990s
Zhang et al., 2004	Eastern China	3	1961–2000	Annual	?	1960s–1990s
Che et al., 2005	China	64	1961–2000	Annual	?	1960s–1990s
Stanhill and Kalma, 1995	Hong Kong (China)	1	1958–1992	Annual	?	1960s–1990s
Stanhill and Cohen, 2005	EE.UU	106	1961–1990	Annual	Jordan, M-M, and Foster	1960s–1990s
Cutforth and Judiesch, 2007	Canada	7	1957–2005	Seasonal and Annual	CSSR	1950s–1990s
Liang and Xia, 2005	China	192	1961–2000	Seasonal and Annual	CSSR	1970s–1990s
Soni et al., 2012	India	12	1971–2005	Annual	CSSR	1970s–1990s
Qian et al., 2006	China	537	1955–2000	Annual	?	1970s–1990s
Liepert and Kukla, 1997	Germany	8	1964–1990	Annual	CSSR	1960s–1990s
Stanhill, 1998a	Ireland	8	1955–1995	Seasonal and Annual	CSSR	1960s–1990s

[a]The suggested role of aerosols in each case is also shown.
[b]Campbell–Stokes sunshine recorder (CSSR), Maring–Marvin thermometric recorder (M-M), Foster photoelectric Sun switch sunshine recorder (Foster), Jordan photographic recorder (Jordan), and Eko sunshine duration sensor (Eko).
[c]Within the study period, the subperiod showing discrepancies between trends of SD and trends of clouds and/or solar radiation.
[d]Symbols indicate different degrees of trend in each case: nonsignificant (=), large positive (+ +), positive (+), large negative (− −), and negative (−).
[e]Discrepancies are attributed to the direct effect of aerosols (DE) and/or the indirect effect of aerosols (IE). Discrepancies are explained without the introduction of aerosols (NO).

Since the long-term series of SD and total cloud cover are widely available, the relationships between both variables have been studied since the early twentieth century. The two quantities are considered to be mutually complementary (see section 3.1): an increase (decrease) in TCC is generally accompanied by a decrease (increase) in SD (see, for example, the high correlation between SD and cloud cover at 1165 stations worldwide in *Wang et al.* [2012b], Figure 10). Therefore, a large literature record relating these two magnitudes and studying their annual, seasonal, and monthly evolution is available [e.g., *Fox*, 1961; *Angell et al.*, 1984; *Angell*, 1990; *Jones and Henderson-Sellers*, 1991; *Weber*, 1994; *Palle and Butler*, 2001; *Curto et al.*, 2009; *Matuszko*, 2011]. But this relationship is not always simple and linear and can be affected by several factors, such as changes in atmospheric transparency resulting from aerosols or changes in the properties of clouds.

The present paper specifically addresses studies that have demonstrated inconsistencies between the trends of TCC and SD. For instance, a decrease (increase) in SD is not always accompanied by an increase (decrease) in TCC (Figure 5); this fact was revealed by several authors [*Morawska-Horawska*, 1985; *Kaiser*, 2001; *Zheng et al.*, 2008; *Sanchez-Lorenzo et al.*, 2009; *Jaswal*, 2009; *Sanchez-Lorenzo and Wild*, 2012; *Kitsara et al.*, 2013; *Wang et al.*, 2013]. Most of these studies have proposed, as one important cause of decreases or increases in SD, an increase or decrease in emissions of carbon and sulfur compounds that scatter and absorb radiation, thus increasing (or decreasing) the turbidity of the atmosphere. *Zheng et al.* [2011] took a step forward; they found a significant declining trend in SD over south China with a nonsignificant trend in TCC and water vapor, while visibility (used as a proxy of aerosol concentration) exhibited a significant

Table 1. (continued)

Author	SD Trend[d]	TCC Trend	LCC Trend	G Trend	D Trend	I Trend	Aerosols[e]
Morawska-Horawska, 1985	−−	−−					D.E.
Kaiser, 2001	−−	−−					D.E.
Zheng et al., 2008	−−	=					D.E.
Sanchez-Lorenzo et al., 2009	−−	=					D.E.
Jaswal, 2009	−−	=					D.E.
Sanchez-Lorenzo and Wild, 2012	++	=					D.E.
Wang et al., 2013	−−	=					D.E.
Zheng et al., 2011	−−	=					D.E.
Kaiser and Qian, 2002	−−	−−					D.E.
Changnon, 1981	−	++					D.E. and I.E.
Kitsara et al., 2013	++	++					D.E.
Li et al., 2011	−−	−−	++				D.E. and I.E.
Xia, 2010	−−	−−	++				D.E. and I.E.
You et al., 2010	−−	−−	++				D.E. and I.E.
Brunetti et al., 2009	++	++					NO
Plantico et al., 1990	−	++					NO
Liu et al., 2002	−−	=					NO
Yang et al., 2012	−−	−−					NO
Power, 2003b	=			++	−−	++	D.E.
Zhang et al., 2004	−			−−	+	−−	D.E.
Che et al., 2005	−			−−	+	−−	D.E.
Stanhill and Kalma, 1995	−	=		−−			D.E. and I.E.
Stanhill and Cohen, 2005	=			−−			NO
Cutforth and Judiesch, 2007	=			−−			NO
Liang and Xia, 2005	−−	−−		−−		−−	D.E. and I.E.
Soni et al., 2012	−−	−−	−−	−−			D.E. and I.E.
Qian et al., 2006	−−	−−	−−	−−			D.E. and I.E.
Liepert and Kukla, 1997	=	=		−−			D.E. and I.E.
Stanhill, 1998a	−−	=		−−	−−		D.E. and I.E.

downward trend. They concluded that the decrease in SD was partially due to the enhanced aerosol loading resulting from anthropogenic activities. Equally, *Kaiser and Qian* [2002] found that a large portion of southeastern China, the region presenting the largest decrease in SD across this country, showed the biggest increases in aerosol extinction coefficient during the 1954–1998 period. Since a downward trend in TCC was also identified [*Kaiser*, 2000; *Xia*, 2013], the increase in aerosol extinction appears to support the theory that aerosol loading plays a major role in decreasing SD.

Wang et al. [2012a] hypothesized that the SD trend in China was affected by TCC, precipitation, and aerosols (quantified through the air pollution index (API)). To isolate the impact of air pollution on SD, *Wang et al.* [2012a] selected clear days (i.e., days with 0 mm of precipitation and TCC ≤10%) and grouped them into two categories: API ≤ 80 (low-medium aerosol loading) and API > 80 (high-aerosol loading). They concluded that aerosols negatively affect SD in China (Figure 6). *Wang et al.* [2012b], analyzing data from more than 1100 stations over the world and filtering the cloud contribution, observed that approximately 58% of all stations had a correlation coefficient greater than 0.5 between SD and the
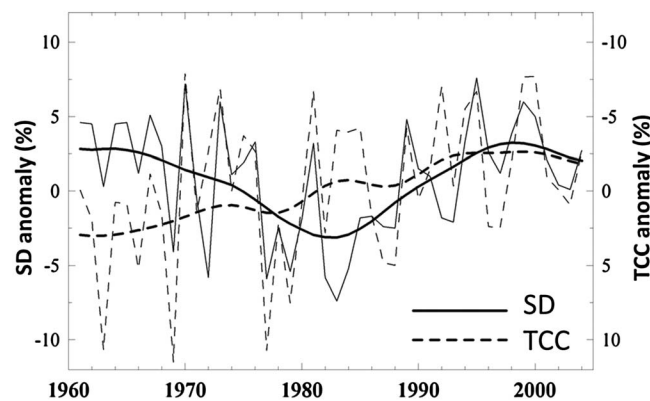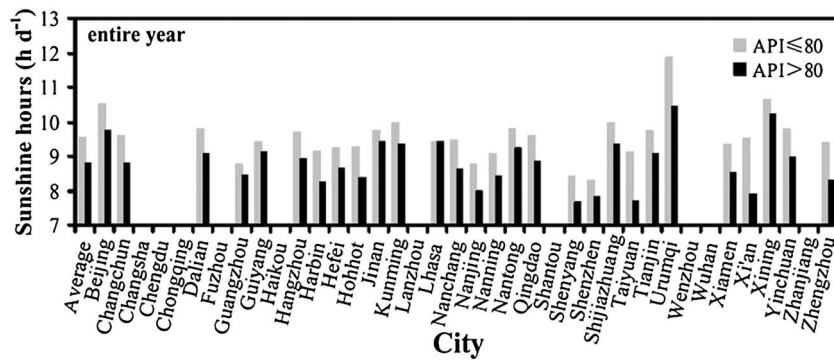

Figure 5. Thin lines represent the annual evolution of both sunshine duration (solid lines, labeled as SD) and total cloud cover (dashed lines, labeled as TCC; note the reversed axis) series for the whole Iberian Peninsula during the period 1961–2004. Thick lines are the smoothed (11 year averaging) evolution. Note that there is a clear disagreement between both series, especially from the 1960s to the mid-1980s. Adapted from *Sanchez-Lorenzo et al.* [2009].

Figure 6. Average daily sunshine hours under conditions with low-medium aerosol loading (air pollution index ≤ 80) and high aerosol loading (API > 80) across China for the 2001–2005 period. Note that the plots are only for clear-sky conditions (i.e., days with 0 mm precipitation and total cloud cover ≤ 10%) and that SD is shorter at times of higher API (high aerosol loading). From *Wang et al.* [2012a].

aerosol optical depth (AOD) for a period of time from the 1980s to the 2000s (AOD is estimated by visibility data, supplemented with satellite data). They also found that some sites showed a low or even negative correlation. At these stations, cloud variability was the determining factor of long-term variation in SD.

SD trends can be better explained by trends of low cloud cover (LCC) than by TCC, as high clouds are more transparent than low and medium ones. For example, *Li et al.* [2011] and *Xia* [2010] found a significant increase in LCC combined with decreases in SD and TCC in China, as well as a higher LCC correlation with SD than with TCC. Both the trends and the correlation coefficients therefore suggest that LCC is the main factor of decreasing SD in their study area. Nevertheless, *Li et al.* [2011] also found a significant correlation between SD and visibility (used as a proxy of aerosol concentration), suggesting a link between SD and aerosols in their study area. *You et al.* [2010] obtained similar results on the Tibetan Plateau; i.e., SD has a significant correlation with LCC and a nonsignificant correlation with TCC. *Xu et al.* [2006] studied the annual variation trend in SD and LCC in Beijing City and its metropolitan area and found that the decline in SD and the increase in LCC are more abrupt in the south peripheral area, where the highest concentration of aerosols occurs. The same behavior was found by *Shi et al.* [2008], who detected areas in southeastern China, where AOD derived from Total Ozone Mapping Spectrometer measurements correlated positively with LCC and negatively with SD. In summary, a decrease in SD appears to be related to the increase detected in LCC. On the other hand, LCC changes can be also linked to indirect aerosol effects.

Some studies do not consider aerosol changes on attempting to explain discrepancies between cloudiness and SD series. *Brunetti et al.* [2009] found significant disagreements in long-term trends of SD and TCC in some subregions of the Greater Alpine Region, but air pollution could not justify the same sign in the TCC and SD, as those subregions were less affected by air pollution. By way of an explanation, *Brunetti et al.* [2009] proposed a poor station density in the above-mentioned subregions. *Plantico et al.* [1990] questioned the homogeneity of the long-term cloud and sunshine records, rather than attributing cloud/sunshine discrepancies to physical factors. Moreover, *Liu et al.* [2002], attempting to explain the observed ~15% reduction in SD in Taiwan from 1960 to 1990, found no significant trend in TCC. They attributed this discrepancy to the subjective nature and the well-known great uncertainty of cloud observations in the detection of trends in the order of 10% (similar to the reduction of SD). Finally, *Yang et al.* [2012] found a decreasing trend of both SD and TCC over the Tibetan Plateau. In light of this paradox, they initially suggested the increase in aerosol loading as a possible cause of the solar dimming over this area [*You et al.*, 2013], but they found that it was not sufficient to explain it. Instead, they proposed that the decrease in SD was mainly due to an increase in water vapor amount and greater cloud cover (due to low air density and strong surface heating) and that there was no need to introduce aerosols.

Trends in solar radiation can also be directly related to SD trends. Based on physical principles, an increase (decrease) in SD must reflect increases (decreases) in direct (*I*) and global (*G*) radiation and decreases (increases) in diffuse radiation (*D*). This means that high positive correlations are to be expected between SD and direct and global irradiation. Some studies explore deviations from these good correlations and attribute the inconsistencies to changes in aerosols.

For example, *Zhang et al*. [2004] and *Che et al*. [2005] found a decrease in SD in China but with a lower rate than the reduction of *G* and *I*. Air pollution was suggested to be the possible cause of these decreases, as it absorbs and scatters solar radiation. According to *Stanhill and Kalma* [1995], the fact that the rate of decrease in SD was lower than the rate of decrease in *G* and *I* indicates that long-term increases in aerosols induce a more significant reduction in the intensity of solar radiation than in its duration.

Furthermore, some articles do not find SD trends despite the existence of G or I trends. For some stations in Germany, *Power* [2003b] found a nonsignificant trend in SD, while the long-term decreases in aerosols were the most likely cause of the observed increases in *G* and *I* and the decreases in *D*. Similarly, *Stanhill and Cohen* [2005] and *Cutforth and Judiesch* [2007] found no long-term SD trend but rather a significant reduction of *G* during the last 50 years of the twentieth century in the United States and on the Canadian Prairie, respectively. They attribute this discrepancy to the different sensitivity of SD and *G* measurements to diurnal changes in cloud cover, and to the influence of other variables such as humidity, without needing to introduce aerosols. For example, an increase in midday cloud cover and air humidity would reduce annual values of *G*, without necessarily affecting SD, whereas the same increases occurring during the morning or evening hours would have the opposite effect. Moreover, *Stanhill and Cohen* [2008] found that the long series of atmospheric turbidity measurements available (used as an index of aerosol content) showed no long-term changes to explain the increase in *G* and SD in Japan, and they proposed that the most likely cause was the change in TCC; however, cloud data were unavailable. Thus, *Stanhill and Cohen* [2008] concluded that the causes of the increase in SD and *G* could not be established unequivocally, given the lack of TCC data.

Meanwhile, *Cohen and Kleiman* [2005] studied the *I* and SD series in Jerusalem under stable synoptic summer conditions, which were characterized by negligible cloudiness during most hours of the day, and found a significant decrease in SD and *I*. Studying hourly SD data, they concluded that in the evening, the Sun cannot be seen with the naked eye before the calculated sunset, whereas the appearance of the Sun came after the previously calculated sunrise. Thus, the length of the day has apparently become shorter during recent decades, due to a hypothetical increase in aerosol anthropogenic emissions. Previously, *Aksoy* [1999] had found the same effect in Ankara around sunset and sunrise as well. In particular, he detected the decreases in SD in each hour of the day that cannot be fully explained by changes in cloudiness, the longest reductions being at sunrise and sunset, i.e., at low solar elevation. Unlike *Cohen and Kleiman* [2005], who associated this change in SD with aerosol loading, Aksoy attributed the trend in SD to the trend in relative humidity, which causes changes in the burning threshold and consequently reduces the hours of SD from sunrise to sunset. However, Aksoy also suggested that the weaker correlations between SD and relative humidity during the winter months may have arisen from aerosol anthropogenic emissions, which show an increase in the winter months.

Thus, both solar radiation and cloudiness data are required to obtain robust results on how aerosols affect long-term SD trends. *Liang and Xia* [2005] observed decreasing trends in SD, TCC, *G*, and *I* over much of China. As the trends in cloud amount and solar radiation were negative and quite similar, cloud amount was not the cause of the decrease in solar radiation. They suggested aerosol loading as the principal cause of the observed decline, due to the rapid increase therein as was revealed by means of visibility data. *Qian et al*. [2006] and *Soni et al*. [2012] observed that some stations in China and India, respectively, presented a decline in *G*, SD, and TCC and also in LCC, so cloud cover changes could not serve as a "universal" explanation of solar dimming; they concluded that changes in the amount and optical properties of anthropogenic aerosols, as well as cloud properties, are the most probable causes of the reduction in surface solar radiation and consequently in SD. It is important to remark that the interactions between aerosols and cloud formation hinder complete separation of their effects on radiation and SD [e.g., *Rebetez and Beniston*, 1998; *Sanroma et al*., 2010], since aerosols may either enhance [*Albrecht*, 1989] or inhibit [*Ackerman et al*., 2000] cloud formation. For example, *Liu et al*. [2002] found that differences between the reductions of SD in urban centers and those of rural areas were substantially smaller than the differences in observed aerosol optical depths among those stations, so *Liu et al*. [2002] suggested that direct scattering by aerosols is not the major cause of the reductions in SD. Indeed, *Stanhill and Kalma* [1995] and *Liepert and Kukla* [1997] had already noted the importance of the effect of aerosol loading on cloud optical depth and on cloud type. *Stanhill* [1998a] could not explain the reduction in irradiance and SD at Valentia (Ireland), as it cannot be attributed to changes in TCC, and he concluded that more investigation was required on variation in cloud cover transmissivity associated with cloud type and/or aerosol load.

Some other authors have analyzed the effect of other meteorological variables on SD measurements. First, *Oguz et al.* [2003], *Yang et al.* [2009a, 2009b], *Zongxing et al.* [2012], and *Wang et al.* [2012a] found that wind significantly influences aerosol loading and, in consequence, SD; on days with strong winds, the aerosol loading was lower than on days dominated by weak wind. Wind direction is also important because winds coming from deserts (dust winds), sea, or industrialized zones could increase aerosol loading and thus negatively influence SD. Second, atmospheric water vapor absorbs solar radiation in some bands and thus can affect SD, as I could fall below the threshold level [*Oguz et al.*, 2003; *Yang et al.*, 2009a; *You et al.*, 2010]. For example, *Van Beelen and Van Delden* [2012] found an increase in SD and a decrease in relative humidity (which can be related to the columnar water vapor content in the atmosphere) in Netherlands. Thus, the decline in humidity might have contributed to the clearing of the atmosphere, causing an increase in SD. A statistically negative correlation between SD and relative humidity was found by *Zongxing et al.* [2012], reflecting the significant influence of humidity on SD: increased (decreased) water vapor decreases (increases) received solar radiation. Finally, *Qian et al.* [2007] commented that a decrease in relative humidity may affect SD positively through reduced absorption and via the aerosol hygroscopic effect (i.e., particles take up water when relative humidity is high, increasing their diameters, and changing their radiative properties [*Tang*, 1996; *Baynard et al.*, 2006]).

4. Conclusions and Future Perspectives

In the present paper, we have reviewed studies addressing the possibility that sunshine duration measurements can be used to detect changes in atmospheric turbidity. According to the reviewed literature, there is an evidence that SD records contain signals of the direct effects of aerosols on the solar beam, and in consequence, SD records can be used as proxy for studying aerosol trends and their radiative forcing. This finding may be crucial in the study of the dimming/brightening phenomenon and its causes.

If the direct effect of atmospheric aerosols on SD records is to be determined, the effects of cloudiness should be previously removed from SD measurements. Despite the fact that aerosol modifies cloud amount and properties, this indirect effect is difficult to quantify by means of SD data. Therefore, the aerosol direct effect has been the only one considered by researchers when quantifying the signal of aerosols with SD data.

Studies showing links between changes in SD and aerosols do not focus upon proving and quantifying the causal relationship. Indeed, the signal of aerosols on SD records is weak, and instruments that measure SD present certain limitations. Moreover, changes in the way of measuring SD affect the homogeneity of the series and lead to errors when evaluating their trends. Moreover, water vapor in the atmosphere is important for two main reasons: first, apart from absorption of solar radiation by the vapor itself, as aerosols are normally hygroscopic, changes in humidity might contribute to the clearing/unclearing of the atmosphere, influencing SD measurements; second, the burning threshold of Campbell–Stokes sunshine recorders may be affected by relative humidity through modification of cardboard burning properties.

Despite all these difficulties, there is a need for further study of SD records, because a new method for determining information on aerosols prior to the 1980s would be very useful and would help us to understand the impact of aerosols on climate. For this reason, some authors have attempted to quantify the direct effect of aerosols on SD data. As pointed out by *Jaenicke and Kasten* [1978], *Helmes and Jaenicke* [1984, 1985, 1986], and *Horseman et al.* [2008], SD recorders do not differentiate a small change in direct solar radiation when it is well above the burning threshold. Therefore, the impact of aerosols on SD occurs particularly just after sunrise and before sunset because of the long distances that the direct beam travels to reach the Earth's surface, so being much more affected by the presence of aerosols. These methods are simple but give only a coarse estimation of atmospheric turbidity and pose the major problem of being affected not only by aerosols but also by water vapor.

Thus, the magnitude of the effect of aerosols on SD is a complex issue, but detailed studies of the burn (in particular during cloudless sunrises and sunsets) provides the possibility to determine variations in turbidity for both short and long time scales and therefore to assess the possible influence of human activities on the Earth radiation budget, with the resulting climatological implications.

Finally, we suggest some further research for further study of the suitability of SD records to detect changes in atmospheric turbidity and aerosols:

1. In order to identify the impact of aerosol on SD records, other variables such as cloud cover and visibility are recommended, especially data available at shorter time scales (e.g., daily or hourly), which can help to confirm the effect of aerosols on SD.
2. To extract information on the trends of aerosol loading, a seasonal or monthly basis is more recommendable than an annual one, because the origins of particulate air pollution can vary along the year. For example, in developed countries, more pollution is usually caused by space-heating-related emissions during winter, whereas photochemical processes dominate during summer.
3. The method described by *Horseman et al.* [2008], which involves extracting sunshine amounts at low-elevation angles from cloudless sunrises and sunsets in order to track changes in pollution, could be improved by using the measures of width of burn in CSSR cards [e.g.,*Wright*, 1935; *Lally*, 2008; *Horseman et al.*, 2013; *Sanchez-Romero et al.*, 2012, 2013], as this provides a way to create a time series of solar irradiance and atmospheric aerosol loading metrics reaching back over 120 years from the present day. Long-term records of SD cards are available at some historical meteorological stations, such as the Blue Hill Observatory in Milton, Massachusetts (U.S.), where CSSR cards have been stored since the mid-1880s (Mike Iacono, personal communication, 2014). However, when these methodologies have been validated, they could be applied to long-term series of SD data without analysis of the original burnt cards. A limited number of sites with a few years of burnt cards would be sufficient for the validation.

In conclusion, when the threshold drifts inherent to sunshine recorders have been well characterized and when the length and widths of the burning can be determined in an objective manner, with the use of automated digital image processing techniques, we will likely be able to evaluate the historical aerosol content from SD records, which should be validated by comparing these estimates with atmospheric aerosol concentration derived from surface or satellite observations.

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