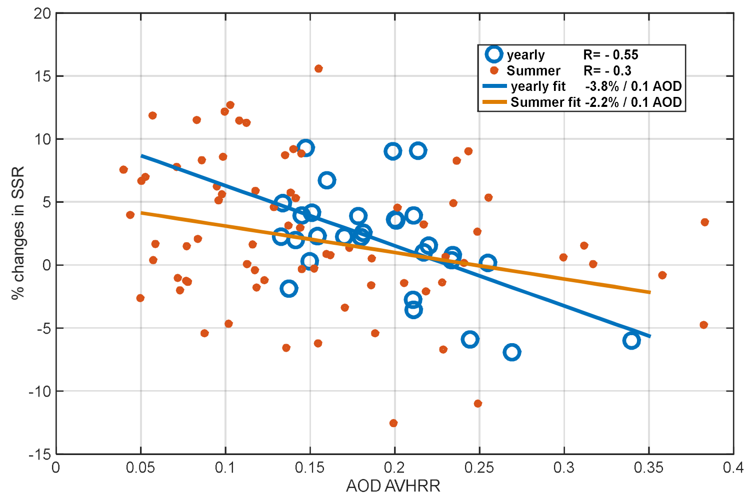
We would like to thank Dr. Tanaka for his fruitful comments.

ALL sky vs AOD

All sky SSR measurements and AOD from AVHRR have been used in order to find the AOD effect in all sky data.For yearly AOD and SSR averages from 1981 to 2009 a correlation coefficient of -0.55 was calculated with a rate of SSR reduction per 0.1 units of AOD equal with -3.8%.For monthly based comparisons, all months revealed a correlation coefficient of -0.2 with a rate of -1.5% /0.1 AOD with better results for summer and Autumn months (-.30, -2.2%/0.1 AOD and -0.30, -1.5%/0.1 AOD)



- The abstract states that a decrease of 2.9%/decade in SSR from 1910 to 1940. I wonder why the trend from this particular period is selectively highlighted within the extrapolated period of 1900-1952. Table 2 indicates a small increase of 0.04%/decade from 1900 to 1952, which is clearly different from above.

We have altered the abstract including:
Very small (0.02%) changes in SSR from 1900 to 1952, including a maximum decrease of 2.9% per decade in SSR from when taking in to account the 1910 to 1940 period, assuming a linear change in SSR.

- Also in the abstract, I could not find where the difference of 4.5% comes from. Table 2 indicated approximately 3.1% but for a slightly different period. The winter period shows the largest change, unlike what is stated in the abstract.

The 4.5% comes from figure 6 as an average of the difference of the 12 months.
Adding the trends of 1953-82 and 1983-2012 gives a slightly different result because mathematically these two individual percentages are calculated using de-seasonalized data using different (for the two periods) mean month values.

- Page 2, Line 21: Regarding the discussion on SSR changes in polluted and pristine areas, I believe that this is still an issue of controversy but two recent studies (Imamovic et al. 2016; Tanaka et al. 2016) showed otherwise, which can be reflected to this statement.

Reference to these studies and corresponding discussion has been added.

– Page 3, Line 2: Fix the citation style.

Style has been fixed

- Page 3, Line 6: Remove "explain".

Extra word has been removed.

- Page 3, Line 22: Figure 1 of (Ohmura 2009) also makes a clear case for this statement.

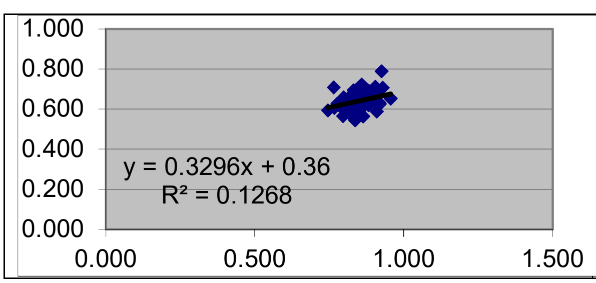
Reference to this work has been added.

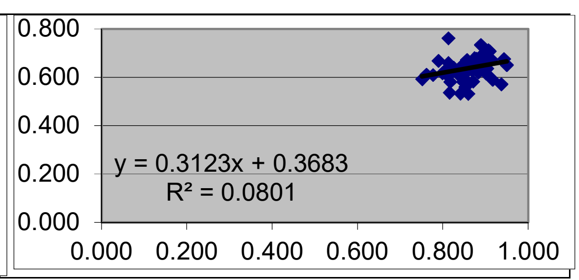
- Page 4: Somewhere in the text (not necessarily in this page), the discussion could touch on aerosol-clouds interactions to acknowledge that the two factors (aerosols and clouds) are not completely mutually exclusive in explaining SSR trends.

Added sentence in page 2:"However, due to the aerosol-cloud interactions and the aerosol indirect effect on SSR, the two factors (clouds and aerosols) are not completely mutually exclusive in explaining SSR changes."

- Page 11, Line 14: I am trying to speculate what causes the weak correlation in summer. The paper cites small ranges of variables in summer as a reason for weak correlation, but how exactly do the range affect R2 values? Later in the paper (Figure 12), the number of cloudless days in summer is generally large, compared in other seasons. Could the number of cloudless days influence the correlation level?

The weak correlation is probably caused by the very low variability of the SDu/SDmax and the SSR/SSRmax ratios. Below an example for July and August correlations Where in XX' axis is the SDu/SDmax and in the YY' axis is the SSR/SSRmax. Large number of cloudless days in the summer is exactly the reason for this low variability. So the calculated Ångström factors for monthly based analysis, based on this example can not be used as only a 12.6% and 8% of the variability of the reconstructed (1900-1953) Julys and Augusts could be explained using these method.

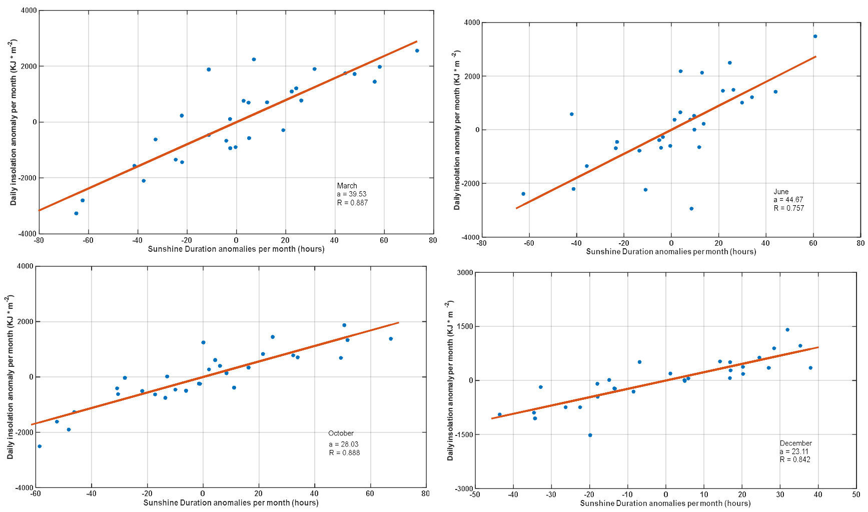




In the initial submission we have used the Ångström related formula in order to calculate SSR and SD related functions. This method includes the theoretical SSR and SD maximum values that insert an uncertainty for such calculations. After the reviewer's comment we decided to replace this method with the one used more frequently for such data series and is described with detail in Sanchez-Lorenzo and Wild, 2012. One additional reason to test this method (as mentioned also in the paper) was the fact that monthly based calculated SD to SSR conversion functions had high uncertainty, linked with the very small SD/SDmax absolute variability especially for summer months.

In this new approach (Sanchez-Lorenzo and Wild, 2012) we did not use SSR and SD theoretical maxima in order to normalize the two factors, but monthly anomalies of SSR and SD have been used for a common measuring period and then the monthly coefficients of the regression of SSR and SD anomalies were used in order to reconstruct the 1900-1953 time series.

The regression statistics of these monthly based SSR and SD anomalies analysis showed much better results from the Ångström method. As an example (and included in the new manuscript) statistics and graphs are shown below.



month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
a	22.47	34.99	39.53	46.65	57.88	44.67	51.87	46.23	34.28	28.02	27.32	23.10
R	0.842	0.895	0.887	0.840	0.799	0.757	0.773	0.572	0.812	0.888	0.916	0.842

According to A. Sanchez-Lorenzo and M. Wild, 2012, the correlation coefficients, here in the range of 0.75 to 0.91 can explain 65% to 82% of the variability of the SSR monthly anomalies. This additional verification analysis shows that the method used in this work is in accordance with important already published results. (e.g.A. Sanchez-Lorenzo and M. Wild, 2012) that have been analysed 17 stations with very long term SDU series.

After having calculated the reconstructed series with this method we have compared the yearly and monthly SSR deviations with the ones calculated with the Angstrom method using the yearly functions (initial submission). The results in yearly basis for all 1900-1953 period differ at a maximum by 1%.

The agreement of these two results shows that in the case that SD measurements in the past have no particular quality issues, then SSR can be reconstructed with the 65-82% explained variability already mentioned.

Finally we have decided to keep the new method on the revised document and include the (yearly based) Ångström results as a verification. The inclusion of this method had a direct impact on all related figures 2, 3, 4, 5, 8 and tables describing trends that include the 1900-1953 period. As already reported the differences were small but still all the plots and tables have been replaced with the new ones calculated based on the Sanchez-Lorenzo, Wild method.

- Page 12, Line 10: "light grey" should be "light blue" from what I can see from the figure.

The colour has been described properly.

- Page 13, Line 1: Separate "late1930's" into two words.

Suggested change has been edited.

- Page 13, Line 7: It may be useful to break up the 1900-1952 period into two because the text discusses the trend till late 1930s and the trend that follows separately.

We think that the current period break up into 1900-1952-1983-2012 periods is already a bit of a mix up for the reader. The basic idea behind this was that the 1900-1952 period is simulated SSR and the 1983-2012 two times 30 year measurement periods that could be also compared with each other. Figures like 5 and 7 could be used to retrieve any SSR % change for any time window and there can be readers that could be interested in a very specific period during these 112 years of reconstructed & measured SSRs.

- Page 13, Line 11: Remove comma after 2012.
Comma has been removed.

- Page 14, Lines 10-13: It needs to be specific which region it refers to. The trend of global anthropogenic BC emissions during 1910-1950 does not decline but rather levels off

(Lamarque et al. 2010).

J.-F. Lamarque et al.: 1850–2000 gridded anthropogenic and biomass burning emissions 7

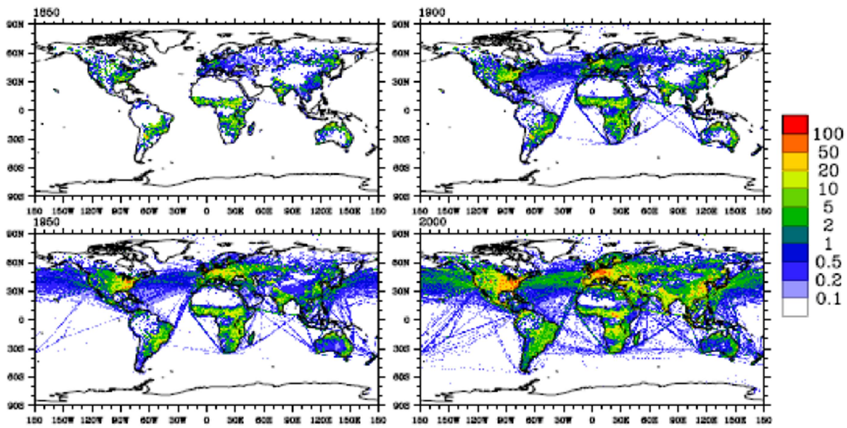


Fig. 4. Total annual emissions (anthropogenic, shipping and biomass burning) of NO_x (Tg(N)/year) for 1850 (top left), 1900 (top r 1950 (bottom left) and 2000 (bottom right).

The sentence has been changed to:
"Nevertheless, early dimming and brightening periods have been reported in Stanhill and Achiman (2016). The results can be partly supported by trends in anthropogenic black carbon (McConnell et al., 2007; Lamarque et al., 2010) and biomass-burning (Lamarque et al., 2010) emissions in Europe."

- Page 15, Line 7: Remove comma after Figure
Comma has been removed.

6. - Page 16, Line 3: Is the left panel of Figure 7 essentially same with Figure 5? If so, the left panel does not have to be shown as it is redundant.

The left panel in figure 7 is linked with figure 5 as it represents a sub period. Mathematically it is not the same as part of figure 5 as de-seasonalized monthly and yearly SSRs have been calculated only for the provided (fig. 7) and not total (fig. 5) period. But we agree with the reviewer that essentially they are the same so we deleted figure 7a but included the discussion on this figure on the relative paragraph.

- Page 18, Line 9: "non significant" needs to be connected by hyphen

Hyphen has been added.

- Page 20, Line 11: the SSR line should be "black" rather than "blue".

The colour has been described properly.

- Page 22, Line 7: Would there be any possible explanation why only the clear-sky SSR trend in winter is negative? A similar result was obtained for the all-sky SSR (Table 2).

There is no straight forward explanation for this negative winter trend. There are various aspects related with the seasonal trend calculation for wintertime such as:

- Wintertime clear sky statistics include more uncertainty due to the more frequent presence of clouds and the fewer clear sky points available.
- Clear sky changes and trends are linked with aerosol changes. For Athens area absolute AOD values for winter are minimum compared with other seasons.

- Page 22, Line 25: The discussion on visibility can be part of the discussion, not the conclusion. Visibility has not been brought up since the introduction.

The section has been transferred to the discussion section and only the conclusions of this discussion has been left to the conclusion section.

- Page 23, Line 3: “drown” should be “drawn”.
Typo has been corrected

Referee 3

We would like to thank the reviewer for his/her comments. We have tried to answer as detailed as possible.

Page 3, line 4: AOD trend of 0.05 per decade: in what period?
The period 1979-2009 was added in the text

P. 3, l. 9: "This attenuation may be much larger ..."
This sentence has been restated

P. 5, l. 17: "Maximum error on the daily integral SSR..."
This sentence has been restated

Table 1 and associated discussion: there are relatively long periods between instrumental changes (up to 6 years). How the radiometers were calibrated prior to 1992? Which was the reference scale? Were the instruments compared with the old one before substitutions? Was the occurrence of instrumental drifts checked?

All instruments used in the study were accompanied by the calibration certificate of the manufacturer in the radiation scale used at that time. This was primary used for calibrating the instruments. For the presented time series the information on homogenization methods and major corrections made, is getting sparser going back in time. For instrument changes till 1968 there are only reports (no actual data) that refer to overlapping periods of measurements for two instruments with just comments on overlapping related corrections and no information on the magnitude of corrections. The two Eppley pyranometers have been calibrated 2 times during the period 1975-1986 in the World Radiation Center in Davos Switzerland and the calibrations acquired have been used for processing the data.

There are several publications that have used the 1954-1983 time series of NOA that are reported in the following comment.

More information on the instrumentation in use and details about temperature correction:

For Solarigraph Gorezynski pyranometers, used during the period 1953-1959, it is well known (Coulson, 1975; Robinson, 1966) that no temperature compensation was provided in these instruments. The temperature coefficient was about 0.0015-0.0020 per 1^{o}C in the sense of decreasing sensitivity with increasing temperature. This theoretical rate have been used.

Two Eppley 180 pyranometers have been used in NOA between 1960 and 1973 period. Solar radiation measurements published in the official NOA's Bulletin were temperature compensated improving considerably the performance of these two pyranometers. According to Drummond (1965), accuracies of the order of ±2-3% are attainable for daily summations of radiation with temperature compensated Eppley 180 pyranometers. That uncertainty does not include instrument drifts with time (if any).

All Eppley 8-48, 8-48A and PSP pyranometers, used in NOA for solar radiation measurements from 1974 until now, are equipped with a built-in temperature compensation circuit. According to Coulson (1975) and Hulstrom (1989), Eppley 8-48 pyranometers provide a signal which is independent of temperature to within ±1.5% from -20 to +40°C. Also, Eppley PSP pyranometers present a reduced temperature dependency of sensitivity on ambient temperature of ± 1% from -20 to +40°C.

On the other hand, the uncompensated Eppley 180 pyranometers are subject to a significant dependence of sensitivity on the temperature of the instrument. The sensitivity decreases with increasing temperature by between 0.05 to 0.15% per 1°C rise in the temperature over the range -50 to +40°C (Coulson, 1975; Robinson 1966).

P. 6, l. 6-12: the application of different data selection criteria, with the addition of quality checks based on the diffuse irradiance, may potentially influence the results of the trend analysis, Did the author check that this is not the case?

The addition of the diffuse irradiance synchronous data in the quality control procedures in the recent years, of course, decreases the uncertainty of the daily, monthly SSR calculations, compared with the one for past years. However, there is no specific scientific evidence or hint in the calculations that points towards a systematic overestimation or underestimation of the SSR data when not using the diffuse radiation "controls". Such an improvement on the quality control methods is in line with most of the long term SSR data series that have been published worldwide.

P. 7.l. 1-2: as far as I understand, the night-time dark signal was subtracted from daytime measurements. This procedure reduces but does not eliminate the thermal offset of the instruments. It must be taken into account that the different types of radiometers display a quite different thermal offset; in general, this is much larger for PSP than for 8-48 or 180° pyranometer. Thus, a systematic overestimate of the SSR in daytime, up to 3-4 W/m2, is possibly present in the data after 1989. This may potentially produce an artificial positive trend in SSR in the recent year; at least, an additional uncertainty should be considered in the trend analysis. Did the authors take into account this effect?

The effect has been tackled based on the temperature corrections that has been mentioned in the first comment but mostly by the overlapping comparisons and homogenization during the 1986 (last) change of instrumentation. Comprehensive studies (Solar and Infrared Radiation Measurements, Energy and the environment, by Frank Vignola, Joseph Michalsky, Thomas Stoffel, CRC Press, 2016) have pointed out to the PSP related thermal offset issue. In our case the subtraction of the night time dark signal (more specific the mean of the previous and next night signal was subtracted for a specific day) reduces at least in half the problem. However, in order to answer to the reviewer question, this (remaning 1-2 W/m^2 ?) was not considered in our analysis as another part of this have been tackled through the overlapping measurements/homogenization procedures. The possibility of such an uncertainty is mentioned in the new manuscript.

Concerning the first three comments on the instrument performance for the period 1954-1986 we decided to include a list of publications that have used the SSR presented time series for this period:

- Macris, 1959, have used the 1954-1956 SSR measurements to identify the relationship of SSR and sunshine duration.

- Katsoulis and Papachristopoulos, 1978, have used the NOA SSR data from 1960 to 1976 in order to calculate SSR statistics for daily, seasonal and yearly solar radiation levels for Athens, Greece.
- Notaridou and Lalas, 1979, have been used the 1954-1976 SSR data from NOA in order to verify an empirical formula on global net radiation over Greece.
- Flocas, 1980 have used the 1961-1975 SSR time series to compare them with sunshine duration data for the period.
- Kouremenos et al., 1985 have used the SSR data from 1955-1980 to correlate changes with various atmospheric parameters.
- Zabara, 1986 have used the 1965-1980 time series to verify a developed method that calculated monthly solar radiation.
- Katsoulis and Leontaris, 1981, have used the 1960-1977 data to verify tools describing the solar radiation distribution over Greece.

We have included these references in the new document.

The reference to these studies does not automatically means that the 1954-1986 data do not include uncertainties related with the calibration frequency and quality control, but they are mentioned as a proof of the scientific data quality for the given period, based on the work of various solar radiation related scientists.

Moreover: there are some rapid changes in the series that may require additional scrutiny; some of these seem to be in correspondence or close to the dates of the radiometers' replacements (e.g., possibly in 1960, 1968, 1973). This seems even more evident in figure 8 from the de-seasonalized monthly mean SSR. Was the presence of step-changes in the series, mainly in corrispondence with instrument replacement, checked?

Specifically on the changes mentioned and linked with the instrument changes.

1960: We have to rely on the reports that state that the two time series have been homogenized, as we did not find other than the officially published NOA bulletin data, that did not include overlapping measurement periods.
1968: Same as 1960.
1973:The Eppley pyranometer, type 8-48A, was recalibrated in 2004 by the Laboratory of Meteorological Device Calibration (LMDC, Psiloglou, 2016), with an uncertainty of 1.2%. It presents a decrease in sensitivity with a rate of 0.5% per year.

General comment

As mentioned in the uncertainty analysis section, the uncertainty for at least the first two decades of measurements is higher than the one in the last 30 years. According to the log books of instrument users, comparisons of two instruments, before exchanging from the one to the other, has been performed and data have been corrected. As reported we have added a large paragraph describing this aspect and also pointed out in the abstract and conclusion sections this issue. (In the end of this comment.)

One of the "problems" with the time series is that compared with other European long term SSR studies the changes of SSR during brightening and dimming periods are visible but to a lesser extent. This, in addition to the fact that the uncertainty of the 1953-1982 period is higher compared to the last 30-years period leads to the conclusion that the small long term changes at least for the first period (1953-1982) could be within the instrument's uncertainty.

However, small drifts and enhanced uncertainties are linked worldwide with past data of such series, as instrumentation and quality control procedures are improving with time and information quality on the initial decisions is more frequently and scientifically reported.

In order to indirectly try to tackle the question of the data quality of the 1954-1983 period including instrument changes and unaccounted possible instrument drifts, compared with the 1984-2012 period, we investigated the following:

We included in figure 8 a shaded area representing a possible (one direction) "uncertainty" based on reconstructing the 1954-1983 series using: the 1984-2012 measured SSR data and the sunshine duration data for 1954-1983. The reconstruction has been performed in the same way as the 1900-1953 one (new method, see comment below). The one direction "uncertainty", points out possible drifts and instrument exchange related uncertainties. However, that does not mean that we believe more the reconstructed through sunshine duration 1954-1983 series than the actual SSR measurements. If this was the case, we would have decided to present a 1983-2012 high quality measuring period and a 1900-1983 reconstructed one. There are various of such papers published quite recently (small measuring period compared with the reconstructed one: Garcia et al.,, 2014; [1992-2013 measurements reconstructed back to 1933] and Anton et al., 2017 [1887-1950 using radiative transfer modelling]) while in our case we would like to try to use the best way possible the historical SSR measurements of NOA during the 1954-83 period.

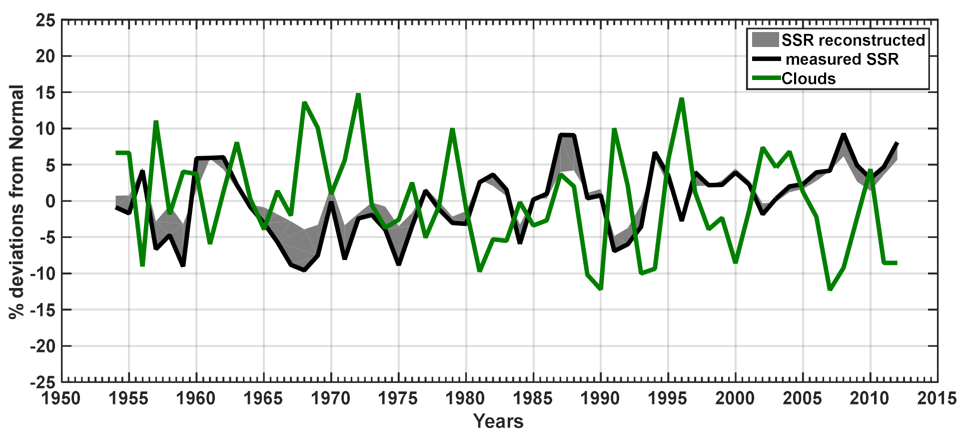


Figure 8 including the reconstructed series and an one direction uncertainty estimation of

In order to investigate more on the reviewer question about the link of data quality of the exchanging instruments period to the calculated trends we have calculated:

A reconstructed SSR series for 1954–1983 using the 1984-2012 SSR-SD calculated functions has been performed, exactly the way the extrapolation has been performed for the 1900-1953 period.

So this case is similar to as if we had only measurements after 1984 and we extrapolated back to 1900 using the functions calculated from the 1984-2012 period.

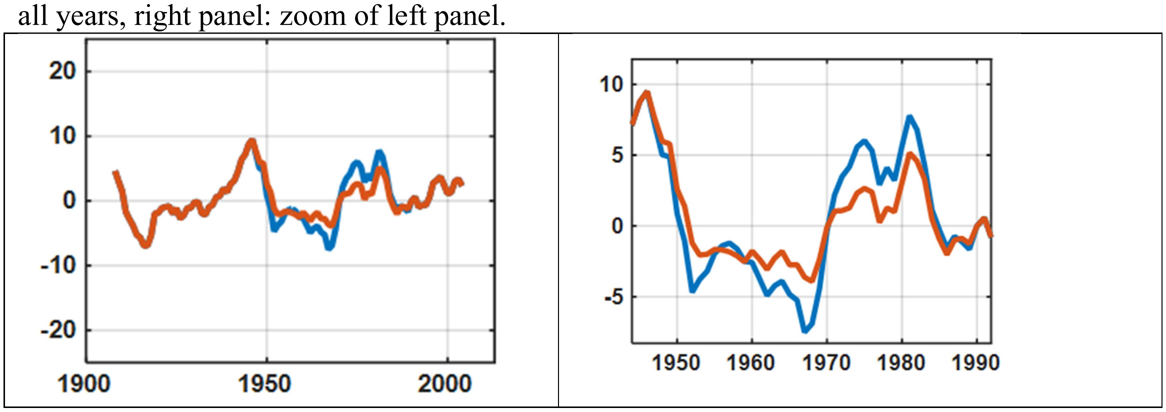
We do not believe that the reconstructed data for the 1954-1983 period are more accurate than the measurements but this investigation has been performed in order to see what would be the effect on the calculated trends if we replace the 1954-1983 SSR measurements with reconstructed SSR values.

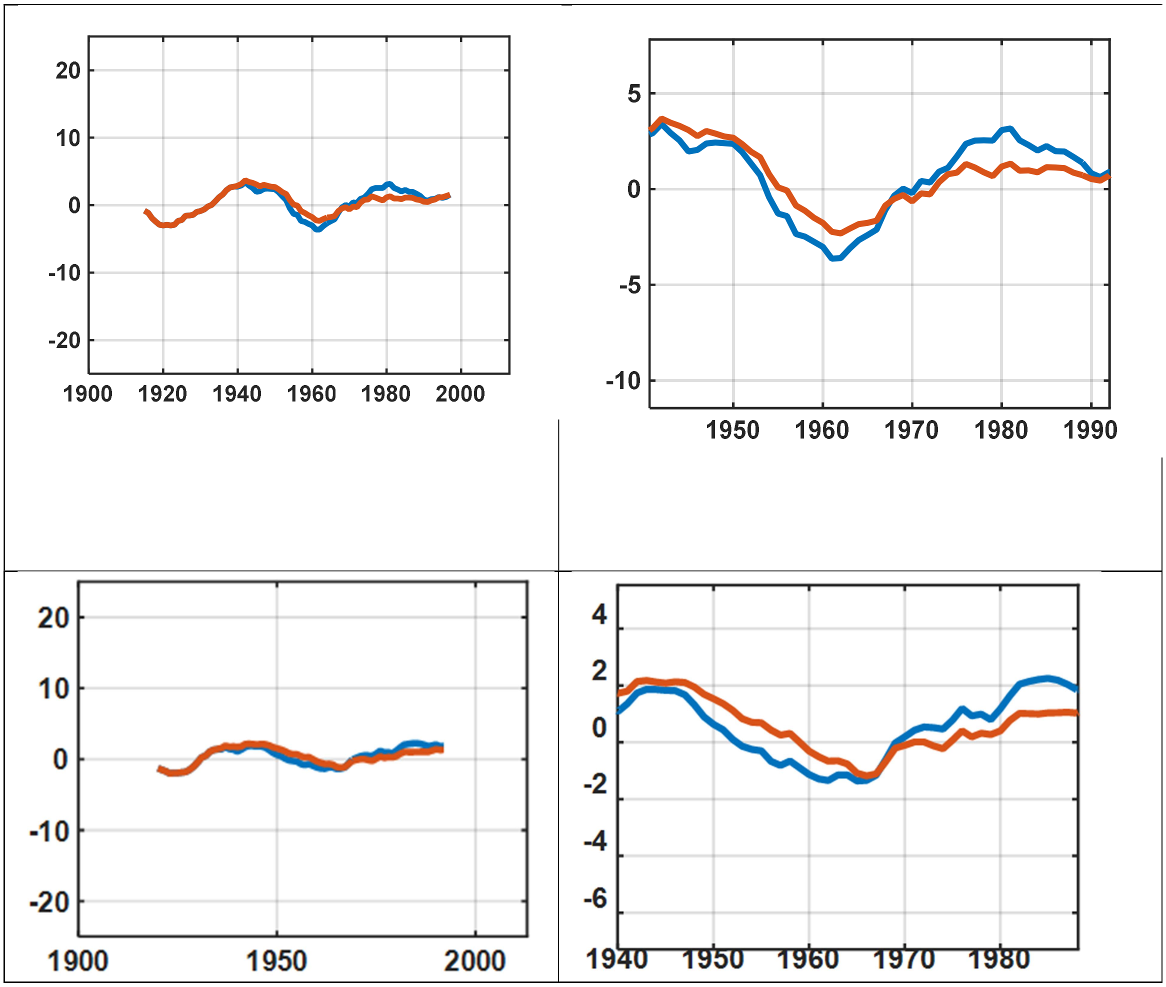
Following, we present the trends per decade using the 15, 30 and 40 year windows. Blue curves represent the trends calculated using the measured 1954-1983 SSR data (they are the same as the ones presented in figure 4 of the paper) and orange lines the trends with the reconstructed (1900-1983) data. (figures a: the full series, b; a zoom in the 54-83 period). Here we have to comment again that the reconstruction method is different than the one of the submitted paper (see comment below).

Differences for the 15 year window differences on the calculated trends outside the 54-83 period are less than 1%, with maximum differences at the late 60's 1-3%. For the 30 year window maximum differences are in the order of 1-2%., while for the 40 year window, maximum differences are less than 1%.

This particular exercise cannot be defined as an uncertainty assessment on the 1954-83 measurements, as reconstructed data cannot be used as a reference. Moreover, as SSR is much more sensitive than SD especially with respect to aerosol optical depth changes. So, in locations where the number of cloudless days is relatively high SD reconstruction tends to "smooth" the SSR variability, however the opposite can be said in cases with constant cloudiness where SD hours in certain days could be close to zero or zero while SSR is never zero.

Figures: Up to down; trends per decade using 15, 30 and 40 year windows, blue line: using the measured 1954-1983 data, orange line: using the 1954-83 reconstructed data, left panel: all years, right panel: zoom of left panel.





Based on all the discussion above we have decided to include a summary of most of the mentioned information on the new version of the manuscript. In addition, a paragraph mentioning:

‘When trying to use such long term series it is evident that the data quality differs as instruments have been improved, quality assurance and quality control procedures have been standardized and finally the information flow on the day to day instrument performance issues are much more frequent in the recent years. More specific for the Athens station, after 1986 the instruments were calibrated or checked with a reference instrument in a yearly basis to identify changes in the calibration and drifts. As reported, the addition of diffuse irradiance measuring instruments provided the opportunity to improve also minute based measurement quality. Before 1986 the instruments reported in table 1 have been used. According with the log books there has been always a certain overlap when changing from one instrument to another. Reports mention that there were instrument drifts that has been corrected with no further information from 1953 to 1970. Instrument overlaps after 1986 were used to eliminate possible instrument related offsets. However, instrument differences (e.g. thermal offset of PSP instrument compared with 8-48 pyranometer, Vignola et al., 2016) theoretically could have an effect in the order of 1-2 W/m^2 on the series continuation. In addition, the inclusion of diffuse radiation in the quality assurance tests after 1991 could have a major improvement on the newest data compared with the old ones. However, there is no hint that the improvement in quality control could have a systematic impact on SSR measured changes compared to the past, other than higher uncertainty on the integrated (monthly, yearly) values, by inclusion of “problematic” SSR minute or small period measurements that did not pass the quality controlled tests. For the 1953-1986 time series there is a number of publications that have been using the SSR-NOA time series. More specific: Macris, 1959, have used the 1954-1956 SSR measurements to identify the relationship of SSR and sunshine duration. Katsoulis and Papachristopoulos, 1978, have used the SSR data from 1960 to 1976 in order to calculate SSR statistics for daily, seasonal and yearly solar radiation levels for Athens, Greece. Notaridou and Lalas, 1979, have used the 1954-1976 SSR data in order to verify an empirical formula on global net radiation over Greece. Flocas, 1980 has used the 1961-1975 SSR time series to compare them with sunshine duration data for the same period. Kouremenos et al., 1985 have used the SSR data from 1955-1980 in order to try to correlate their changes with various atmospheric parameters. Zabara, 1986 has used the 1965-1980 time series to verify a developed method that calculated monthly solar radiation. Katsoulis and Leontaris, 1981, have used the 1960-1977 data to verify tools describing the solar radiation distribution over Greece. Finally, the percentages of errors reported in table 1 are not directly linked with possible instrument drifts, that can impact the SSR time series analysis. So results of measurements before 1986 have to be used with caution and accompanied by a report on the different level of uncertainties of the past and recent data.

In addition we have added a text summarizing the effect of the real and reconstructed 1954-1983 data to the overall calculated trends per decade based on the above figures. New figure 8 seen above in this comment was also replaced the current one mentioning the periods with possible instrument drifts.

Concerning the abstract and conclusions we have added sentences on the data quality:

Absolute year to year changes in SSR for the 1954-1983 measurement period have to be treated with caution as they can include uncertainties related with instrument exchange and undetected/uncorrected instrument drifts. However, long term (e.g. 25 year windows and more) calculated trends could only partly affected by such uncertainties. If reconstructed series can be used as a hint for such drifts/steps, their level is not higher than 2% and only for very specific periods.

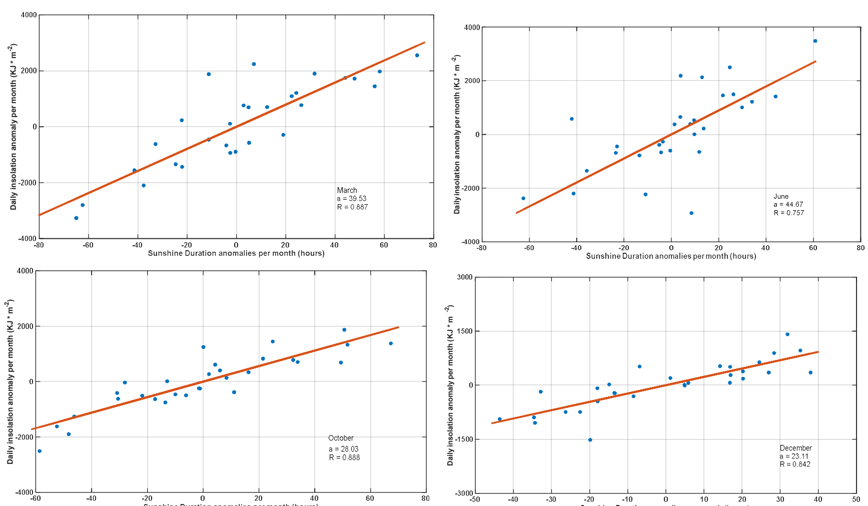
P. 9, l. 31: how are SSR max and SDmax calculated?

Maximum values of SSR and SD in use were theoretical extraterrastial and astronomical value accordingly. The uncertainties related with these parameters and their effect on the reconstruction method was tested by a new reconstruction method used. In this new method such assumptions were not included. See specific comment below.

P. 12, figure 4: estimated SSR/SSRmax values (figures 2 and 3) show a typical 10- 20% spread around the fitting line. This is expected, since the used relationship takes into account only cloud duration. All other effects (most of the aerosol direct effects, as well as most changes in cloud properties) can not be reproduced by the method. What is the uncertainty associated with these SSR estimates? Was this uncertainty considered in the trend analysis? Can these data be reliably used for trend analysis?

In the initial submission we have used the Ångström related formula in order to calculate SSR and SD related functions. This method includes the theoretical SSR and SD maximum values in order to work that insert an uncertainty for such calculations. After the reviewer's comment we decided to replace this method with the one used more frequently for such data series and is described with detail in Sanchez-Lorenzo and Wild, 2012. One additional reason to test this method (as mentioned also in the paper) was the fact that monthly based calculated SD to SSR conversion functions had high uncertainty, linked with the very small SD/SDmax absolute variability especially for summer months.

In this new approach (Sanchez-Lorenzo and Wild, 2012) we did not use SSR and SD theoretical maxima in order to normalize the two factors but monthly anomalies of SSR and SD have been used for a common measuring period and then the monthly coefficients of the regression of SSR and SD anomalies were used in order to reconstruct the 1900-1953 time series.
The regression statistics of these monthly based SSR and SD anomalies analysis showed much better results from the Ångström method. As an example (and included in the new manuscript) statistics and graphs are shown below.



month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
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R	0.842	0.895	0.887	0.840	0.799	0.757	0.773	0.572	0.812	0.888	0.916	0.842

According to A. Sanchez-Lorenzo and M. Wild, 2012, the correlation coefficients, here in the range of 0.75 to 0.91 can explain 65% to 82% of the variability of the SSR monthly anomalies. This additional verification analysis shows that the method used in this work is in accordance with important already published results. (e.g. A. Sanchez-Lorenzo and M. Wild, 2012) that have been analysed 17 stations with very long term SDU series.

After having calculated the reconstructed series with this method we have compared the yearly and monthly SSR deviations with the ones calculated with the Angstrom method using the yearly functions (initial submission). The results in yearly basis for all 1900-1953 period differ at a maximum 1%.

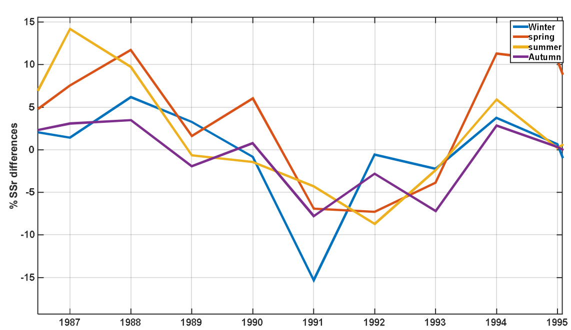
The agreement of these two results shows that in the case that SD measurements in the past have no particular quality issues, then SSR can be reconstructed with the 65-82% explained variability already mentioned.

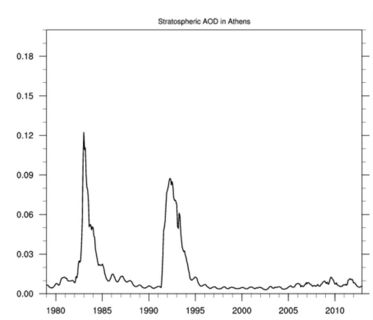
Finally we have decided to keep the new method on the revised document and include the (yearly based) Ångström results as a verification. The inclusion of this method had a direct impact on all related figures 2, 3, 4, 5, 8 and tables describing trends that include the 1900-1953 period. As already reported the differences were small but still all the plots and tables have been replaced with the new ones calculated based on the Sanchez-Lorenzo, Wild method.

Also, it is surprising that no significant signals of large volcanic eruptions (Agung in 1963, El Chichon in 1984, Pinatubo in 1991) are present in figure 4. A small SSR reduction in the early 90's, possibly related with Pinatubo, appears in figure 8; however, the minimum during 1990's in fig. 4 seems too late to be ascribed to Pinatubo (whose effect lasted for up to 2 years). Is there a possibe explanation?

El Chichon date is linked with a 7% drop from 1983 to 1984 and a 5% increase in 1985. So we think that that it is at least partly visible when looking at the whole time series where year to year observations are more linked with cloud conditions.

Figure 4 in the previous manuscript included a wrong shift of one year that is why it did not exactly matched with figure 8. In addition, smaller differences raised from the fact that as normal period was the 1900-2012 while in the new document the 1984-2012. This was corrected, in addition to the (small) changes that were introduced with the change to the Sanchez – Lorenzo reconstruction method.
Here is a zoom of the 1987 – 1995 SSR for the four different seasons, Together with the stratospheric aerosol load calculated based on the ChArMEx AOD (Nabat et al., 2013) series.





We believe that the ~6% drop from 1990 to 1991-1993 shown for all seasons is a hint of the effect of the eruption on SSR data for the Athens station. However, as shown in figure 8 cloudiness for 1991 is also high, while is much lower for 1992 and 1993. Combined with the stratospheric AOD figure, seems that 1991 related decrease is also related with cloud increase while 92 and 93 one with the Pinatubo related aerosol effect.

P.13, l.2: the graph also shows a clear decrease during 1950's.
This sentence has been restated

P.13, l.3: shows
Typo has been corrected

P. 13, table 2: why were these periods chosen? P. 13, l.
The periods have bene chosen based on the data retrieval (reconstruction or measurements) (1900-1952 period and 1953 to 2012). And then measurement period has been divided in two 30 year periods in order to try to compare the two (figure 6).

19-22: does the trend determination and its statistical significance take into account uncertainties?

A sentence has been added:
This has been extensively discussed in a comment above.
We have added:
It has to be noted that the trend determination and its statistical significance does not take into account measurement or SSR reconstruction related uncertainties, which are different for the different periods.
In addition to the new figure 8 and related comments and the comments on the reconstructed-actual 54-83 series related trends.

P. 14, figure 5: this figure does not seem to support the choice of the periods used in table 2 for the trend calculations.

Yes the periods have been chosen based on different criteria (see comment *P. 13, table 2, above*. Figures like fig. 5 can be used for any reader to draw his conclusion on any SSR change under any time window.

P. 17, fig. 8: the units for yearly mean SSR and total cloud cover are missing in the graph. The evolution of the yearly mean SSR does not seem to be coherent with the annual series of de-seasonalized SSR in figure 4 (the minimum in early 1990's does not seem to coincide with the minimum in mid 1990's in figure 4; the minimum in 1970 in figure 4 appears earlier in figure 8). Is there an explanation for that?

The units are % deviations from normal for both SSR and clouds.
See comment above for differences in previous fig. 4 and 8.

P. 18, l. 7: "presence of" may be removed

It has been removed.

P. 18, l. 8: figure 12 and the related discussion suggest that there is a long-term change in the number of cloudy days. Conversely, no significant change in the annual mean cloud cover appears. May this be taken as an indication of changes in cloud properties or distribution?

Yes that is really interesting. We added a paragraph:

“Differences on the ratio of cloudless days shown in figure 12 and on the almost constant cloud octa variability shown in figure 8 is partly attributed to the different definition of a cloudless day that is based on the cloud radiative effect for fig. 12 and on observation of cloud percentage in the sky for fig. 8. However, this can also be an indication of changes in cloud properties (e.g. change in optically thin clouds that could have small radiation effect but are marked as cloudy conditions from the observer).”

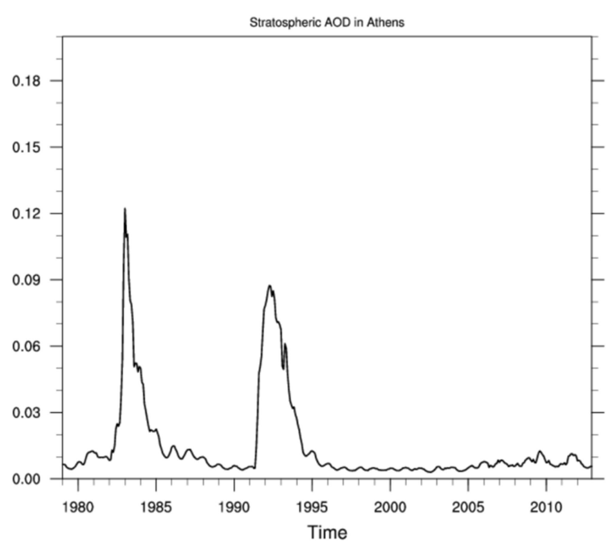
P. 18, l. 20: it may be emphasized that the clear sky selection criterion eliminates cases with high aerosol optical depth.

We agree. A sentence has been added.“It may be emphasized that the clear sky selection criterion could possibly eliminate cases with very high aerosol optical depth.

P. 21, l. 4-5: apparently, there is no stratospheric aerosol contribution in the ChArMEx AOD dataset. The large volcanic explosions are important events with an expected impact on SSR, and datasets which include these cases should be used. Please, explain more clearly what is the meaning of "..uses the trend and not the interannual variability which is not included in the global model that was used".

The ChArMEx AOD (Nabat et al., 2013) accounts for tropospheric aerosols only, and does not include stratospheric AOD coming from large volcanic explosions. We agree that such explosions have an important impact on SSR. We have provided the evolution of stratospheric AOD in Athens (below), where the two main peaks are respectively due to El Chichon and Pinatubo eruptions. These two peaks are possibly associated with decreases in SSR in Athens (Figure 11 in the paper).

Concerning the second part of the comment, the trend in the ChArMEx AOD comes indeed from a global climate model which has no nudging towards reanalysis. Consequently it was impossible to deduce the interannual variability from this model, that is the reason why the ChArMEx AOD only accounts for the trend in AOD due to the decrease of anthropogenic emissions.



Stratospheric AOD over Athens

P. 21, l.12-13: a change of almost a factor of 2 in the frequency of cloudless days seems to be non marginal. No evident effect appears on SSR in figure 8. However, trends in table 3 are calculated in periods separated around the years with minimum number of cloudless days. May part of the trend change in the two periods due to the long-term change of cloudless days/cloud properties (see also comment to p. 18, l.8)?

Based on figure 12 there is a negative change in the number of cloudless days from 1970 to 1985and a positive one from 1985 to 2000.Figure 7a (now deleted after the recommendation of reviewer 2) shows this effect in total (cloudless plus cloudy) SSR. It shows an ~ +5% change for the first period (e.g. blue line year 1977-78) and a ~0% change for the second period (e.g. blue line year 1992-93).

However the problem is more complex as for cloudless days the AOD plays an important role and for cloudy days the cloud radiative effects also play a role always as a function of solar elevation which determines the SSR measurement absolute value.

P. 22, l.25-P.23, l. 9: this discussion seems not fully consistent with the conclusions of the paper. For instance, Founda et al (2016) show that visibility is strongly related with AOD; and the paper highlights a possible role of aerosols in affecting SSR.

The discussion on the visibility now transferred as last paragraph of the conclusions

(Purely mathematically speaking) "In this work and any other work that use SD to reconstruct SSR time series, reconstructed SSR is purely driven by actual sunshine duration changes. Founda et al., 2014 has presented the change of the SD since 1900. Using the measurements data we can calculate the % deviations for SD since 1900. These are in accordance with the reconstructed SSR. More or less the decrease of SSR from 1910 to ~ 1940 and the increase afterwards till ~1950 is also shown in SD. (Founda et al., 2014).

The visibility related study by Founda et al., 2016 shows the visibility variability since 1931. So a first difference is that the first 30 years are missing. A comparison of the SSR results in Athens with visibility observations since 1931 (Founda et al., 2016) did not show any correlation among SSR and horizontal visibility. For the first part of the common dataset (1930-1959) the visibility decline is accompanied with a SSR increase. However from 1950 till today visibility shows a monotonical decrease. The steep visibility decrease from 1931 till the 90's is not accompanied by a relative SSR or SD decrease excluding individual sub-periods.

As already reported, simulated SSR is driven purely by changes in sunshine duration, in this case the SD variability in founda et al., 2014 is almost constant after 1950 so SD also, can not be linked with the visibility reported decrease. Studying the literature for similar cases, similar conclusions have been drawn by Liepert and Kukla (1997) showing an SSR decrease over 30 years of measurements accompanied by a visibility increase and no significant changes in the cloud cover conditions, in Germany. This Athens SSR vs visibility relationship can be partly explained by the fact that: SSR and visibility have different response on cloud conditions, water vapor and rainfall and also by the fact that visibility is affected by aerosols only in the first few hundred meters above the surface, while SSR is affected by the columnar AOD, which in the case of Athens can be significantly different due to aerosol long-range transport in altitude (e.g. Saharan dust; Léon et al., 1999)."

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Long-term series of surface solar radiation at Athens, Greece

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Abstract

We present a long-term series of solar surface radiation (SSR) for the city of Athens, Greece. The SSR measurements were performed from 1954~~3~~ to 2012, and before that (1900-1953~~2~~) sunshine duration (SD) records have been used in order to reconstruct monthly SSR. Analysis from the whole dataset (1900-2012) mainly showed: Very small (0.02%) changes in SSR from 1900 to 1953, including a maximum decrease of 2.9% per decade in SSR ~~from~~ when taking in to account the 1910 to 1940 period, assuming a linear change in SSR. For the dimming period (1955-1980), a -2% change per decade has been observed, that matches various European long-term SSR measurement related studies. This percentage for Athens is in the lower limit, compared to other studies for the Mediterranean area. For the brightening period (1980-2012) we have calculated a +1.5% per decade which is also in the lower limit of the reported positive changes in SSR around Europe. Comparing the 30-year periods (1954-1983 and 1983-2012) we have found a difference of 4.5%. However, measurements of the first 30 year period are associated with higher uncertainties than the second period, especially when looking at year to year changes ~~.~~ The difference was observed for all seasons except winter. Using an analysis of SSR calculations of all sky and clear sky (cloudless) conditions/days, we report that most of the observed changes in SSR after 1954 can be attributed partly to cloudiness and mostly to aerosol load changes.

1 Introduction

In the past decades surface solar radiation (SSR) and the transmission of the atmosphere have been of increasing interest because of the related impacts on climate. Most of the energy in the Earth-atmosphere system is introduced by solar radiation as it provides heating, which creates pressure gradients and ultimately wind, as well as it triggers water, carbon and oxygen cycles through evaporation and photosynthesis. These processes define the climatological conditions, and changes of incoming solar radiation rapidly affect the energy balance (Wild et al., 2015). Interest on the solar radiation changes has also been raised after the development of solar energy applications, which are continuously growing in number over the recent years. Changes in SSR have been recorded over the last century and can be caused either by natural events such as volcanic eruptions or human-related activities, mainly in polluted regions (Wild, 2016). At larger scales (thousands of years) changes in SSR, might have been caused by changes in the Earth's orbit and Sun solar output (Lean, 1997; Ohmura, 2006).

Systematic continuous measurements of SSR were established in the middle of the 20th century at selected meteorological observatories. Solar variations have been investigated in several studies using ground based SSR measurements from various monitoring networks worldwide (e.g., Ohmura, 2009) and also by satellite-derived estimations (e.g. Kambezidis et al., 2010). Overall, most of these studies (Gilgen et al., 1998; Noris and Wild ,2009; Wild, 2009 and 2016 and references therein) have reported a worldwide decrease of solar incoming radiation in the period 1960-1985 (known as dimming period), followed by an increase (brightening period) thereafter. These changes ~~were larger~~ reported to be higher in more polluted and urban areas but have also been recorded in isolated regions such as the Arctic (Stanhill, 1995) and Antarctica (Stanhill and Cohen 1997). ~~More~~ Other recent studies have ~~tried to distinguish~~investigated the effect of urbanization on global brightening and dimming, ~~the local scale changes in polluted urban environments in the same time~~ and found no marked differences among urban and rural SSR time series ~~proved the existence of these trends independently of local sources~~ (Tanaka et al., 2016) and Imamovic et al. (2016) ~~investigated the correlation among these trends and urbanization proxies and found non-significant linkage~~. Changes in atmospheric transmission due to variations in cloudiness and aerosol concentration are the main factors to be investigated in order to determine the possible causes of such trends in SSR (Wild, 2009). However, due to the aerosol-cloud interactions and the aerosol indirect effect on SSR, the two factors (clouds and aerosols) are not completely mutually exclusive in explaining SSR changes.

The cloud and aerosol radiative effects on solar radiation variations over the past decades have been investigated by numerous studies during the last years. The inter-annual variations in cloudiness is crucial for studying SSR time series, but its decadal variability is not always connected with the widespread dimming and brightening effects (Wang et al., 2012; Wild, 2016). Aerosols play significant role in incoming radiation, by scattering and absorbing light and by acting as cloud-condensation nuclei. Over the 20-year dimming phase (from 1960 to 1980) and the 15-year brightening phase (from 1990 to 2005), it was found that the aerosol effects (direct and indirect) played the most important role in SSR variation (Dudok de Wit et al., 2015). Concerning Central Europe, Ruckstuhl et al. (2008) suggested that the brightening phase under cloud-free conditions is in line with decreasing anthropogenic aerosol emissions (Streets et al., 2006). Nabat et al.~~,~~ (2013) using a blending of remote sensing and model products showed that a decreasing Aerosol Optical Depth (AOD) trend of 0.05 per decade has been calculated for Europe for the period of their study (1979-2009). In addition, Nabat et al., 2014 reported that anthropogenic aerosol decline in Europe from 1980 to 2012 statistically explains ~~explain~~ 81 ± 16% of the observed brightening. Overall, changes in anthropogenic aerosol emissions are now considered as the major cause of brightening and dimming effects (Wild, 2016). The gaseous and particulate air pollutants may reduce solar radiation by up to 40% during air pollution episodes (Jauregui and Luyando, 1999). ~~This~~ Aerosol related attenuation is much larger during forest fires, dust events and volcanic eruptions. Vautard et al., (2009), have also reported on a decline of the frequency of low-visibility conditions such as fog, mist and haze in Europe over the past 30 years, suggesting a significant contribution of air-quality improvements

Long-term series of SSR measurements are essential for such studies. One of the main constraints in studying SSR temporal changes is the small number of sites with reliable long-term records, even over areas with high density of stations such as Europe, Japan or the USA. In Europe for example, there are currently less than 80 stations with more than 40-years homogeneous data (Sanchez-Lorenzo et al., 2015), with very few of them operating over Southern Europe. Recently, a high-quality dataset of SSR has been set up over Italy (Manara et al., 2016), but there is still lack of high quality long-term trends in other countries around the Mediterranean Basin.

In addition, even more sporadic measurements are available before the 1950s (Stanhill and Achiman, 2016); the few studies of them have pointed out an SSR increase in the first decades of the 20th century and a maximum around 1950 (Ohmura, 2006 and Ohmura, 2009). This topic is still controversial due to the few long-term series available (Antón et al., 2014). Recently, there have been efforts to reconstruct SSR series in periods with no direct measurements available, using other variables such as sunshine duration (SD), which is available in a large number of sites since the late 19th century (e.g., Stanhill and Cohen, 2005, for USA; Sanchez-Lorenzo and Wild 2012, for Switzerland; Matuszko 2014, for Poland). For example, Sanchez-Lorenzo and Wild (2012) used data from 17 stations in Switzerland, considered SD as a proxy and successfully reconstructed SSR time series since the late 19th century. Thus, they calculated that the variability in SSR monthly anomalies can be explained by SD anomalies in a range of 76%-96%, and a monthly root mean squared error of 4.2 W m^{-2} between recorded and estimated SSR for all-sky conditions and of 5.5 W m^{-2} for clear-sky conditions. Other studies have tried to use pan evaporation as a proxy of SSR, for the first half of the 20th century (Stanhill and Möller, 2008). Kambezidis et al. (2016) used monthly re-analysis datasets from the Modern Era Retrospective-Analysis for Research and Applications (MERRA) and calculated shortwave radiation trends from 1979-2012 for the Mediterranean basin. They reported an increase in MERRA by +0.36 W m^{-2} per decade, with higher rates over the western Mediterranean (+0.82 W m^{-2} per decade).

For the Southeastern Mediterranean, there have been a few studies discussing the brightening/dimming effect. Zerefos et al. (2009) have studied the Ultraviolet A (UVA) changes for the area of Thessaloniki (Greece) from 1984 to 2008. They calculated a 5% positive trend per decade linked to a negative trend in aerosol optical depth (AOD) for the area due to air pollution abatement strategies. The variability of shortwave downward solar irradiance received at Earth's surface over Thessaloniki, Greece for the period 1993-2011 studied the Ultraviolet A (UVA) changes (Bais et al., 2013), where they reconfirmed the upward trend in SSR after 1990 (0.33% per year). They also reported signs of a slowdown in the upward trend in SSR during the beginning of the 2000s.–.Founda et al., (2014) have studied the SD long-term variability over Athens area. They reported a 7% decline in the annual SD from 1951-1982 and a 3% increase from 1983-2011 under all sky conditions. Although, under near clear sky conditions, these percentages are -7% and + 9% for the dimming and brightening periods respectively. Similarly, Founda et al. (2016a) analyzed long-term SD and total cloud cover time series over 15 sites in Greece (the oldest one beginning on 1897). They have shown an increase in SD almost at all stations since the mid-1980s, which in certain areas of Southeastern Greece amounts to an increase of 20 h per year. This increase is not accompanied with synchronous decrease in total cloud cover, possibly evidencing to decreasing aerosols loads, despite the fact that their impact on SD should be lower than on SSR (Sanchez-Romero et al., 2014). Yildirim et al. (2014) have analyzed 41 years of SD measurements in 36 stations in Turkey. They reported a decreasing trend (between 1970 to about 1990) for most of the stations. After 1990 they observed either zero trend variation or a reduction in the decreasing rate of SD for most of the locations. They concluded that the decreasing period might be attributed to human-induced air pollution. Founda et al. (2016b) have investigated the visibility trends over Athens area from 1931 to 2013. They reported a deterioration in the visibility up to 2004 and a slight recovery afterwards, negatively/positively correlated with relative humidity/wind speed and positively correlated with AOD from 2000 to 2013. Finally, Alexandri et al., 2017 studied the spatiotemporal variability of SSR over the EasternMediterranean for the 1983–2013 period using the Satellite Application Facility on Climate Monitoring Solar surfAce RAdiation Heliosat satellite-based product.They reported SSR trend be positive (brightening) and statistically significant at the 95% confidence level (0.2 ± 0.05 W/m^2/year or 0.1 ± 0.02%/year) being almost the same over land and sea.

In this work, measurements of SSR, recorded for 60 years at the center of Athens, are presented. In addition, with the use of the SD measurements that are conducted in Athens since 1900, we could reconstruct the time series of SSR during the first half of the 20th century. These time series (1900-2012) are the oldest, uninterrupted and high quality SSR time series in the SE Mediterranean and one of the oldest in Europe, providing unique information about the variations and trends in the area for the past decades. Time-series of SSR over Athens are presented to try answering questions such as:

Are the dimming–brightening patterns observed in Europe over the past century also observed, at the same extent, over the Eastern Mediterranean?

Is SSR variability during the first decades of the 20th century in Athens in line with the other few locations reporting trends over this period?

Can we verify that anthropogenic aerosols play the most important role on the brightening/dimming observed SSR after 1950 in agreement with other European regions?

2 Data and Methodology

2.1 Data collection and analysis

The SSR data used in this study cover the period from December 1953 to December 2012 and were measured by a series of pyranometers that are mentioned in Table 1. These instruments have been operating continuously at the Actinometric Station of the National Observatory of Athens (ASNOA) (Hill of Pnyx, Thissio), that is located near the center of Athens, Greece (38.00° N, 23.73° E, 110 m above mean sea level). Table 1 presents the instruments and the period of operation, as well as the maximum error ~~in the calculation of the~~on the integrated daily values. References mentioned in Table 1 describe the exact type of errors and uncertainties related to the sensors. In the period 1953-1986, the maximum daily error was about 5%, and 2% afterwards. The spectral response of the sensors is in the range of 285-2800 nm; since 1986 a first-class Eppley PSP pyranometer (WMO, 1983) is operating at ASNOA. Since 1992, frequent calibrations (every two years) have been performed by the NOA's Laboratory of Meteorological Device Calibration (LMDC, 2016) in order to ensure the high quality of measurements. LMDC follows the standard calibration procedure for thermopile pyranometers (ISO 9847, 1992), with exposure to real sunlight conditions and comparison with a standard thermopile (Secondary Standard) pyranometer. LMDC's reference pyranometer, Kipp & Zonen CM21, is regularly calibrated in PMOD/WRC, Davos, Switzerland.

Table 1: History of SSR instruments used at ASNOA. SSR measurements refer to the total solar radiation on a horizontal surface.

	Instrument	Period	Class	Maximum error (daily integral)	Reference	Class	Comments	Resolution
1	Solarigraph GOREZYNSKI	1953-1959	2nd	5%	Coulson (1975)	B	One instrument being used	1 hour
2	Eppley 180º pyranometer (No. 3604)	1960-1966	2nd	5%	Coulson (1975), Drummond (1965)	B	Manual measurements archiving with mvoltometer	1 hour
3	Eppley 180º pyranometer (No. 3604) coupled with a Leeds-Northup recorder, Speedomax, type G	1966-1968	2nd	5%	Coulson (1975), Drummond (1965)	B	Same instrument as #2 with Speedomax recorder	1 hour
4	Eppley 180º pyranometer (No. 3034) coupled with a Leeds-Northup recorder, Speedomax, type G	1968-1973	2nd	5%	Coulson (1975), Drummond (1965)	B	New instrument, same recorder	1 hour
5	Eppley pyranometer, type 8-48 and type 8-48A coupled with a Leeds-Northup recorder, Speedomax, type G	1974-1986	2nd	3-5%	Hulstrom (1989)	B	Type 8-48 and type 8-48A Instruments were measuring alternatively for three years each	1 h
6	Eppley Precision Spectral Pyranometer (PSP)	1986-now	1st	1-2%	Hulstrom (1989)	A	Regular recalibrations. Coupled with a, A/D recorder (Campbell Scientific Ltd. Datalogger, type CR-21X at the beginning	1 min

							until 2003, a CR10X until 2012	

SSR data are processed using a set of quality-control (QC) tests in order to ensure the quality of the data set. The QC procedures include rejection of:

- Measurements for solar elevation angles less than 5 deg;
- SSR values equal to or less than 5 W m^{-2}, during sunrise and sunset, due to the pyranometers' sensitivity;
- SSR values greater than 120% of the seasonally corrected solar constant.

After the initiation of diffuse horizontal radiation measurements at ASNOA in 1991, the following quality criteria were added for rejection:

- diffuse horizontal values greater than the corresponding SSR ones;
- diffuse horizontal values greater than 80% of the seasonally correct solar constant;
- direct-beam solar component exceeding the extraterrestrial solar irradiance.

Also, both total and diffuse horizontal measurements are corrected for the night-time dark-signal offset of the pyranometers.

Mean daily SSR values were calculated from the data set of this study (December 1953 – December 2012); only months with more than 20 days of measurements were considered in the analysis. Over the 60 years of measurements, only three months (January and February of 1998 and March of 2012) did not fulfill this criterion.

'When trying to use such long term series it is evident that the data quality differs as instruments have been improved, quality assurance and quality control procedures have been standardized and finally the information flow on the day to day instrument performance issues are much more frequent in the recent years. More specific for the ASNOA, after 1986 the instruments were calibrated or checked with a reference instrument in a yearly basis to identify changes in the calibration and drifts. As reported, the addition of diffuse irradiance measuring instruments provided the opportunity to improve also minute based measurement quality. Before 1986 the instruments reported in table 1 have been used. According with the log books there has been always a certain overlap when changing from one instrument to another. Reports mention that there were instrument drifts that has been corrected with no further information from 1953 to 1970. Instrument overlaps after 1986 were used to eliminate possible instrument related offsets. However, instrument differences (e.g. thermal offset of PSP instrument compared with 8-48 pyranometer, Vignola et al., 2016) theoretically could have an effect in the order of 1-2 W/m^2 on the series continuation. In addition, the inclusion of diffuse radiation in the quality assurance tests after 1991 could have a major improvement on the newest data compared with the old ones. However, there is no hint that the improvement in quality control could have a systematic impact on SSR measured changes compared to the past, other than higher uncertainty on the integrated (monthly, yearly) values, by inclusion of "problematic" SSR minute or small period measurements that did not pass the quality controlled tests. For the 1953-1986 time series there is a number of publications that have been using the SSR-NOA time series. More specific: Macris, 1959, have used the 1954-1956 SSR measurements to identify the relationship of SSR and sunshine duration. Katsoulis and Papachristopoulos, 1978, have used the SSR data from 1960 to 1976 in order to calculate SSR statistics for daily, seasonal and yearly solar radiation levels for Athens, Greece. Notaridou and Lalas, 1979, have used the 1954-1976 SSR data in order to verify an empirical formula on global net radiation over Greece. Flocas, 1980 has used the 1961-1975 SSR time series to compare them with sunshine duration data for the same period. Kouremenos et al., 1985 have used the SSR data from 1955-1980 in order to try to correlate their changes with various atmospheric parameters. Zabara, 1986 has used the 1965-1980 time series to verify a developed method that calculated monthly solar radiation. Katsoulis and Leontaris, 1981, have used the 1960-1977 data to verify tools describing the solar radiation distribution over Greece. Finally, the percentages of errors reported in table 1 are not directly linked with possible instrument drifts, that can impact the SSR time series analysis. So results of measurements before 1986 have to be used with caution and accompanied by a report on the different level of uncertainties of the past and recent data.

Figure 1 shows the intra-annual variability of SSR at ASNOA based on the measurements from all instruments during the period 1953-2012. Mean daily SSR at Athens ranges between approximately 6 to 27 MJ m^{-2} during the year. Mean and standard deviations were calculated using the 60 year record for each day.

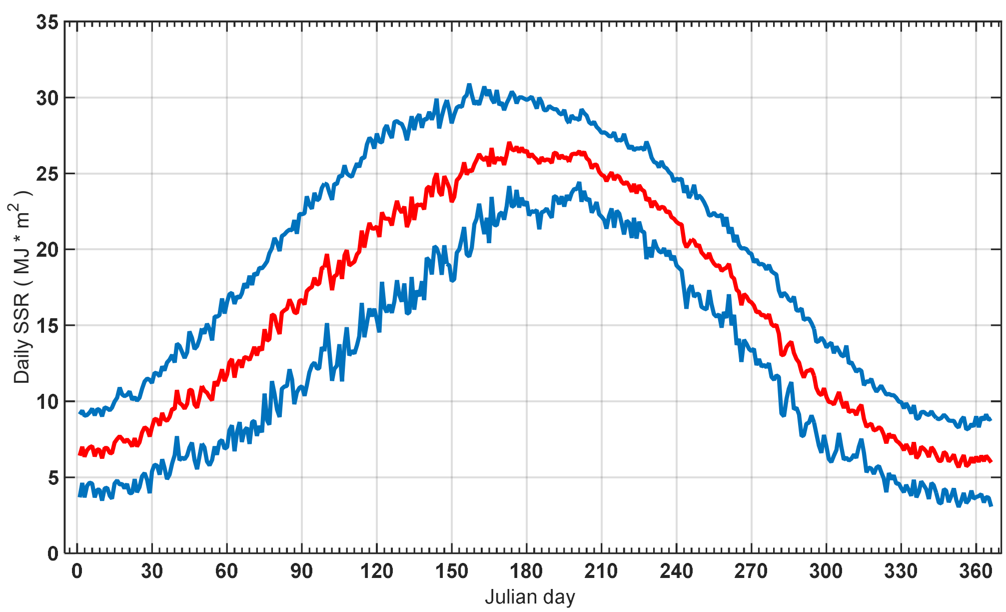


Figure 1. Average Intra-annual variability of Surface Solar Radiation (SSR) at Actinometric Station of the National Observatory of Athens (red), along with the inter-annual variability for a given day (±1 standard deviation, blue), calculated over the period 1953-2012.

The results of figure 1 show the average yearly pattern of SSR at ASNOA. The day to day variability that is shown as "noise" in the plotted blue line comes from the 60 year averaging of each day and is mostly related with the amount of cloudiness for each of the averaged days. Minimum and maximum SSRs at solstices, compared to a cloudless sky aerosol free model, are also related with the highest probability of the presence of clouds during winter months. For the calculation of each of the daily averages the available data points vary from 55 to the possible maximum of 60.

In addition, collocated measurements of SD recorded at ASNOA have been used. According to WMO (2010), the SD during a given period is defined as the sum of the sub-periods for which the direct solar irradiance exceeds 120 W m^{-2}. In Athens, SD has been recorded using classical Campbell-Stokes heliographs (since 1894) and been replaced by electronic instrumentation in 1998 (EKO, MS-091 analog SD sensor). Monthly SD values since January 1900 have been used in this study. A more analytical study of these time series can be found in Founda et al. (2014).

Complementary to this study, cloud-cover observations from the Hellenic National Meteorological Service (HNMS) from 1954 have also been used. These observations are recorded at a site 7 km away from ASNOA. All cloud observations at HNMS are conducted every 3 hours and are expressed in octas.

Concerning the data availability for SSR and SD data: SSR monthly means calculated here have been retrieved from daily calculated SSRs. Over the 59 years (708 months), 98% of the months had none or one day missing, 3 months had from 10-20 missing days and 2 months from 20-30 missing days. For SD, 1931-1940 monthly data have been used taken from the NOA measurement annals. From 1940 on, hourly measurements have been used in order to derive daily and monthly measurements. The SD time series have no gaps with only six missing days during December 1944 (Founda et al., 2014).

In order to examine the AOD impact on SSR, we have used the longest satellite based AOD series available for the area. This is the AOD time series from Advanced Very High Resolution Radiometer (AVHHR). AOD retrievals at 630 nm over global oceans at spatial resolution of 0.1^{o} x 0.1^{o} and one overpass per day, have been used. Data used were downloaded from NOAA Climate Data Record (CDR) version 2 of aerosol optical thickness (Zho and Chan, 2014), and cover the period from August 1981 to December 2009. AVHHR AOD embodies a large variety of uncertainties, including radiance calibration, systematic changes in single scattering albedo and ocean reflectance (Mishchenko et al, 2007). Current dataset radiances have been recalibrated using more accurate MODIS data (Chan et al, 2013). We used daily data at the region around Athens (longitude: 37.5°-38.2°N, latitude: 23.2°-24.4°E) which includes 50 active available (ocean) grid-points. The above region was selected based on data availability on each grid with the distance up to 50 km from ASNOA.

To complement the analysis on the evolution of aerosols, the recent climatology developed by Nabat et al., 2013 has been considered over the period 1979-2012. This product provides monthly averages of AOD at 550 nm over the Mediterranean region at 50 km resolution. It is based on a combination of satellite-derived (MODIS instrument) and model-simulated products (MACC reanalysis and RegCM-4 simulations), which have been selected among many available datasets, from an evaluation against ground-based measurements of the AERONET network. Thus this climatology is able to give an estimation as best as possible of the atmospheric aerosol content over the period 1979-2012. For the present work, the AOD time series over the grid cell of the ASNOA (38.00° N, 23.73° E) has been extracted and is referred to as the ChArMEx data thereafter.

2.2 Clear-sky SSR

For the determination of the clear sky (defined here as the cloudless) days, we have used both the cloud octas and SD data. Daily observations have been used for this analysis. We have defined as a clear sky day each day that fulfills the following criteria:

- the mean daily cloudiness (in octas) should be less than 1.5, and
- the total daily SD should be higher than 90% of its theoretical (astronomical) value.

The procedure for calculating a single mean cloud octa value for each day was the following:

We have first excluded night-time cloud observations; then, we have weighted each observation based on the hour of the observation. Weights have been calculated based on the solar radiation contribution of the specific time slot and day of the month, compared with the daily clear sky SSR integral, of the particular day and month.

2.3 Reconstruction of SSR from SD

We have used the 1900-2012 SD time series in order to extend our SSR time series back to 1900. There are different methods that are used in order to estimate SSR values from SD. In this work we have tried two methods based on the linear regression of SSR and SD (Sanchez-Lorenzo and Wild, 2012). For all-sky conditions the monthly anomalies, obtained as differences from the 1983-2012 mean, of SSR and SD have been calculated. Then for each month a linear regression has been used to estimate the relationship between the SSR and the SD.

Figure 2 shows four out of the total twelve regressions together with its' statistics.

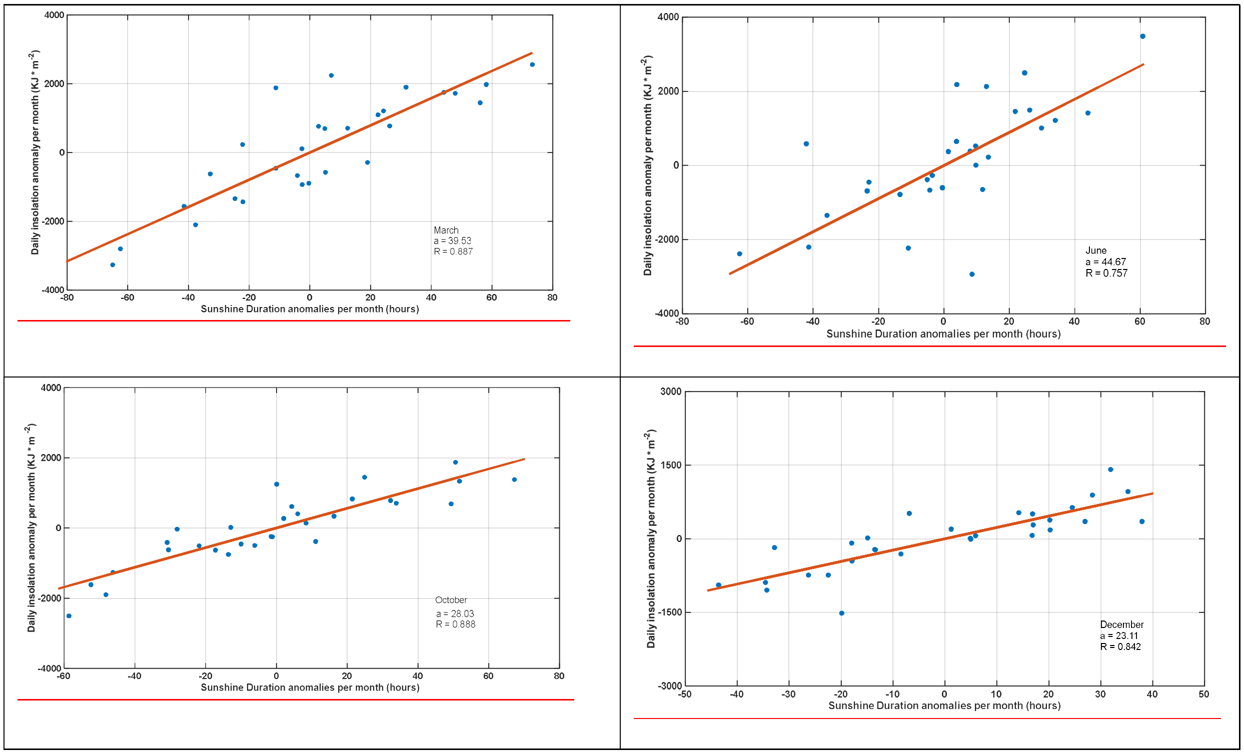


Figure 2. Linear regression of SSR and SD anomalies for March, June, October and December, 1983-2012.

The statistics of the monthly regressions are shown in table 2.

Table 2. Monthly regression statistics of SSR vs SD anomalies.

month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
a	22.47	34.99	39.53	46.65	57.88	44.67	51.87	46.23	34.28	28.02	27.32	23.10
R	0.842	0.895	0.887	0.840	0.799	0.757	0.773	0.772	0.812	0.888	0.916	0.842

where a is the coefficient of the equation SSR= a * SD + b and R the correlation coefficients that vary from 0.75 to 0.91. This implies that the SD monthly anomalies explain between 56% and 83% of the variability of the SSR monthly anomalies. It has to be noted that coefficient b is less than 10^{-3} for all months. In addition to the reported percentages this temporal resolution fitting is more sensitive to inhomogeneities present in the series, as well as local peculiarities and noise.

The second method was based on ~~For that purpose we have used a recent period (1983-2012) in order to derive a function between SD and SSR and a testing period (1953-1982) to verify the validity of this function and results. Finally we applied this derived and tested formula for the 1900-1952 period where SD measurements were available while SSR were not. In order to~~ derivinge a relationship between SD and SSR, ~~we~~ usinged the broadly accepted formula of Ångström (1924):

$$SSR/SSR_{max}=a+b(SD/SD_{max}) \quad (1)$$

where SSR_{max} and SD_{max} refer to the theoretical extra-terrestrial value of SSR and the astronomical value of SD, respectively, while *a* and *b* are constants usually defined monthly. This formula can only be used in large data sets as a statistical approach. That is because for different cloud height, thickness and positioning, the constants can show a large variability (Angell, 1990).

~~3 Results~~

~~3.1 Relationship between SD and SSR measurements~~

~~In order to benefit from the over-century long time-series of SD at NOA we used Eq. (1) for the period of synchronous SD and SSR measurements to estimate constants *a* and *b* for the period 1983-2012.~~ The 1983-2012 period was chosen for determining the SSR vs. SD relationship as mainly SSR measurements have lower uncertainties compared with the 1953-2012 one. We thus calculated a=0.237 and b=0.458 and an R^2 equal with 0.81~~(as derived from the linear equation shown in Figure 2) and used them for validation in the period 1953-1982. Figure 2 shows the correlation of the monthly SSR/SSR_{max} and SD/SD_{max} ratios for the 1983-2012 period, together with the derived coefficients (*a* and *b*) and the coefficient of determination (R^2). In addition, we have included the testing period statistics, together with the coefficients *a* and *b* that can be determined using the whole period (1953-1982), to show that each of the two periods could provide similar results.~~

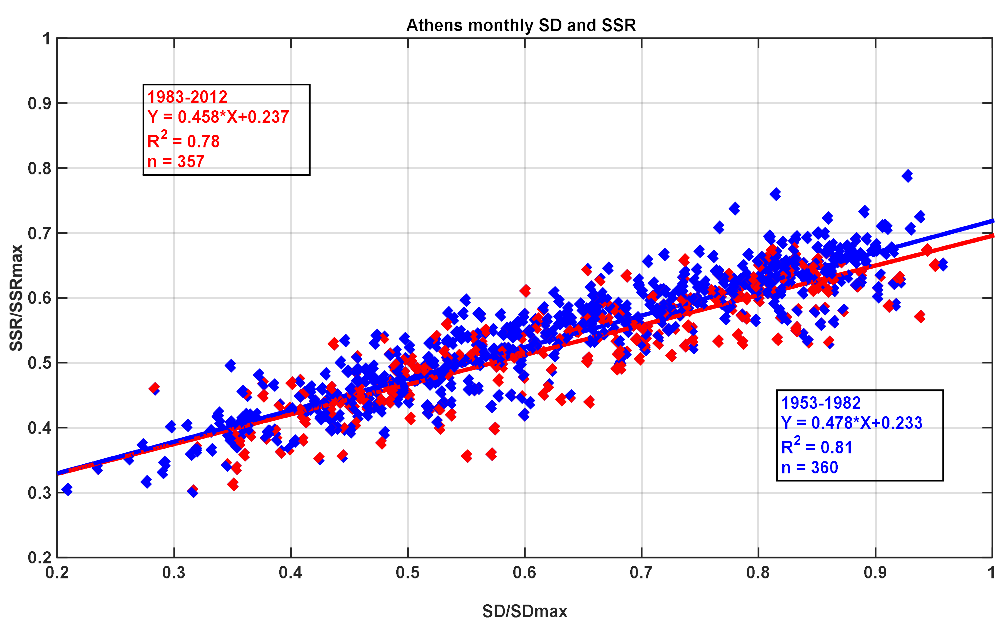


Figure 2. Plot of Y=SSR/SSR_{max} vs X=SD/SD_{max}. Blue dots represent the 1953-1982 period and red ones 1983-2012. The red and blue lines represent the respective linear regression lines. Corresponidng regression relations are given in the inner boxes.

For testing the method we have applied the retrieved coefficients and calculated the SSR for the 1953-1982 period. Comparing normalized simulated with the observed SSR values an R^2=0.79 has been found (see Figure 3).

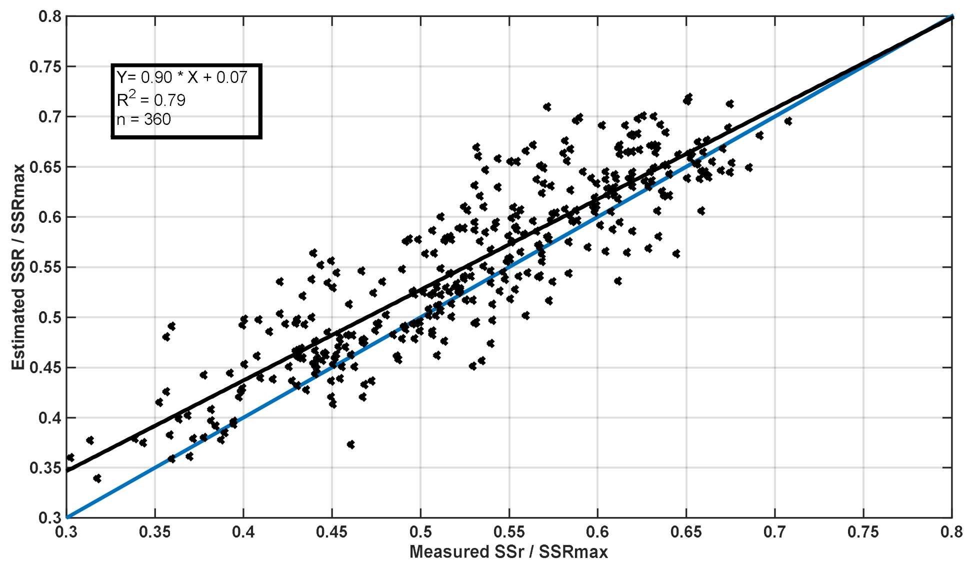


Figure 3. Plot of the ratios of mean monthly SSR values to SSR maximum, of measurements in the period 1983-2012 vs the estimated ratios from Eq (1) for the same period. Blue is the 1:1 and black is the regression line.

We also followed the same procedure to calculate the coefficients of the Ångström formula separately for each month and for each season during the control period 1983-2012. For individual months, calculated SSR/SSRmax vs SDU/SDUmax coefficients of determination ranged from 0.5 to 0.65 for winter months, 0.32 to 0.67 for spring months, 0.47 to 0.53 for autumn months and 0.1 to 0.38 for summer months. So coefficients of determination using the monthly based data were much lower that the first reported method. The low coefficients for the summer period are related with the small range of values of SDU/SDUu max and SSR/SSRmax that are related with the absence of clouds. When we have calculated seasonal based Ångström formulas for winter, spring, summer, and autumn months we have found *a*=[0.22, 0.22, 0.34, 0.27], *b*=[0.48, 0.49, 0.45, 0.34] and R^2= [0.6, 0.74, 0.2, 0.63] respectively.

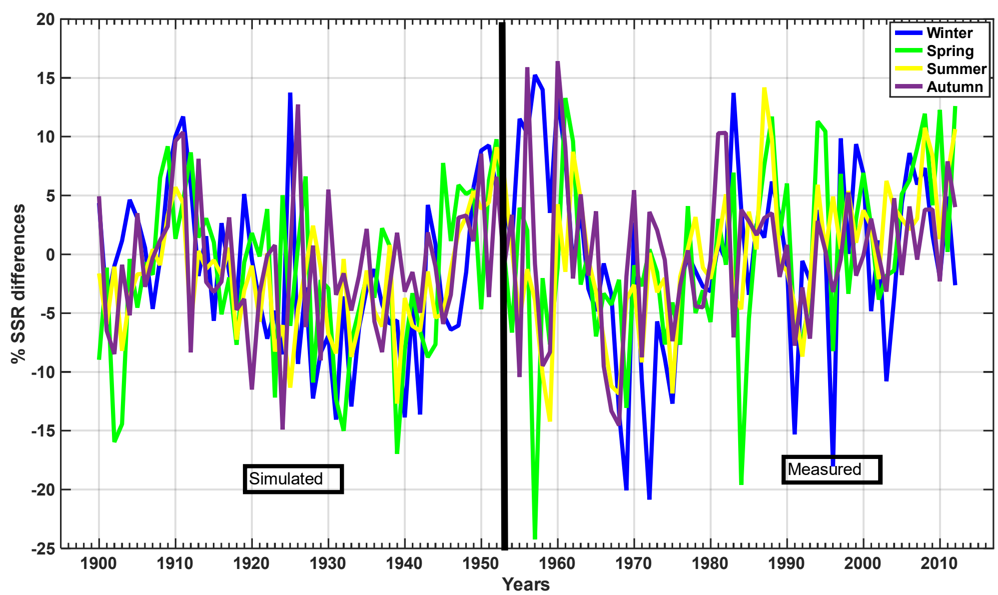
We have used both the monthly regression coefficients from the first method and the yearly based Ångström formulas in order to investigate the impact of the different methods to the SSR reconsruction. Results of the reconstructed SSR yearly values from 1900-1953 showed maximum differences of 1% in the calculated SSR per cent anomalies, while for monthly values the higher differenc was 2%. In order to avoid the use of theoretical normalization values such as SDUmax and SSRmax needed for the second method, we have reconstructed the SSR time series based on the monthly based results of the first method as proposed in Sanched-Lorenzo and Wild, 2012.In order not to include statistical uncertainties introduced from the correlations of individual months and seasons that are reported, we decided to use the Ångström formula derived using all months in the same dataset. Such assumption could introduce a season dependent trend to the extrapolation of SSR back to 1900 but it is considered more safe than using other least trusted seasonal Ångström formulas.

3 Results

3.12 Long-term variations and trends (1900-2012)

Based on the results shownmethods described in in Section 3.12, we have reconstructed monthly SSR from 1900 to 1953. Using the full dataset of reconstructed (1900-19532) and measured SSR (19543--2012) we have calculated the mean monthly SSR values and used them for de-seasonalising the results shown in Figure 34. The de-seasonalizing was determined by: a. calculating the average SSR (SSR_{mi}) for each month (i) out of the 12 months of the given year, for all 1983-2012 years, b. calculating the changes in % in SSR (SSR%(i,y)) for each month (i) of each year (y) as:

$$SSR\%(i,y) = \frac{SSRiy - SSRmi}{SSRmi} * 100$$



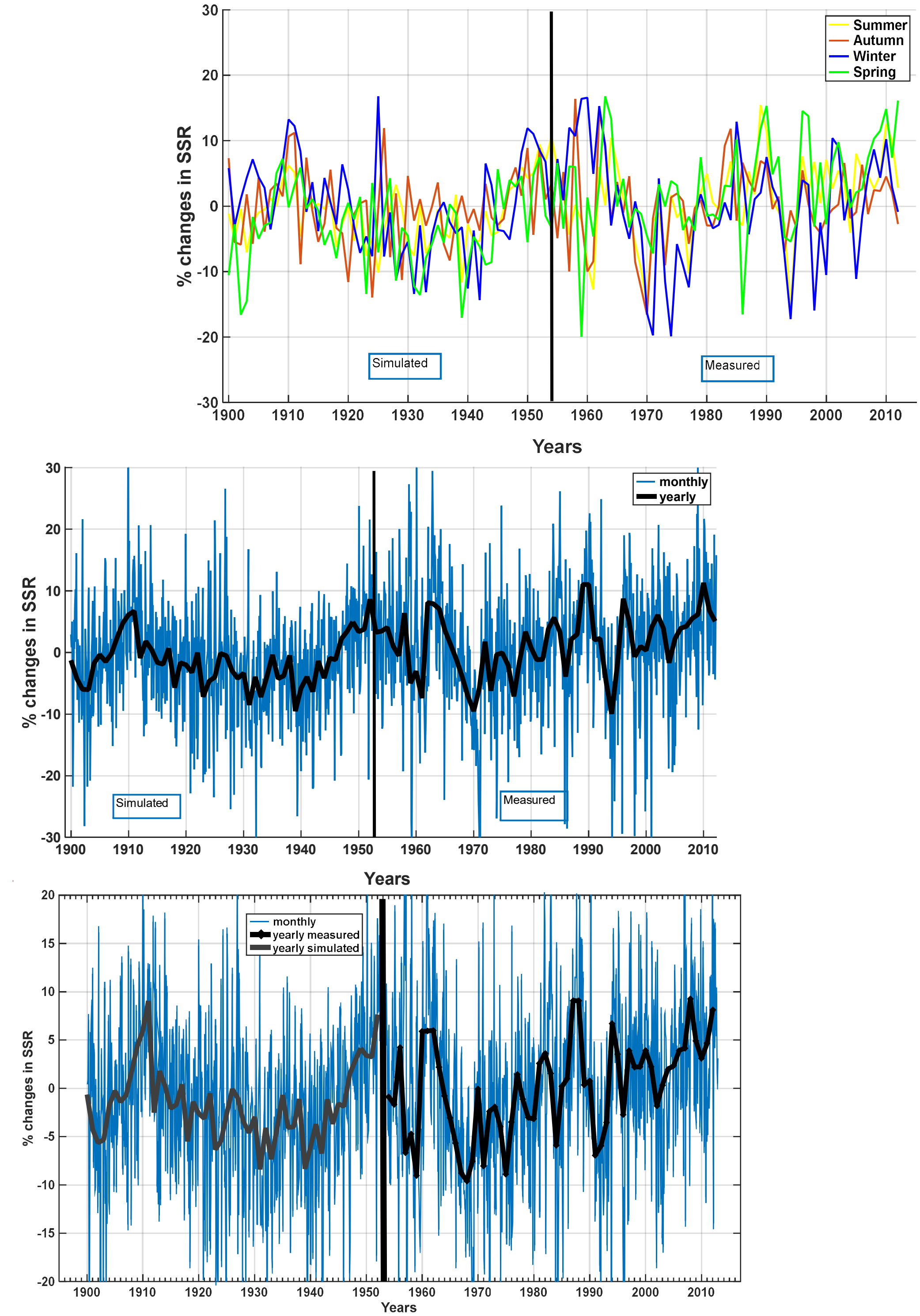


Figure 34. Full time series of de-seasonalised SSR per cent changes (using the 1900-2012 monthly averages). Upper panel: different colors represent seasonal analysis, lower panel: black bold line represents the annual series, and light ~~grey~~ blue line the mean monthly values

According to Figure 4, the month-to-month variation (shown with light grey line) can reach more than 30% in comparison with the mean monthly average of the whole data set. Annual means show a 10%-12% (peak to peak) decrease in SSR from 1910 to late 1930's and then an increase of 12% from 1940 to early early 1950's. The simulated SSR results follow the observed decline of SD reported in Founda et al., 2014, where the same (~15% decrease and increase from 1919 to early 1950's) is shown.

According to Figure 3, the month-to-month variation (shown with light grey line) can reach more than 30% in comparison with the mean monthly average of the whole data set. Annual means show a 10%-12% (peak to peak) decrease in SSR from 1910 to late 1930's and then an increase of 12% from 1940 to early 1950's. The simulated SSR results follow the observed decline of SD reported in Founda et al., 2014, where a decrease from 1910 to 1940's and an to early 1950's is shown.

Subsequently, there is a decrease during the late 1960's 1950's and then a positive change of the order of 20% till today with an episode in the early 1990's that shows low SSR values. Measured SSR in 1991-1993 period differs by 5% compared with the one in 1990.The latest can be linked with the Pinatubo volcanic eruption and its known effect in the SSR (e.g. Zerefos et al., 2012).

Analytical linear trends of each of the sub-periods and for every season are presented in Table 2. It has to be noted that the trend determination and its statistical significance does not take into account measurement or SSR reconstruction related uncertainties, which are different for the different periods.

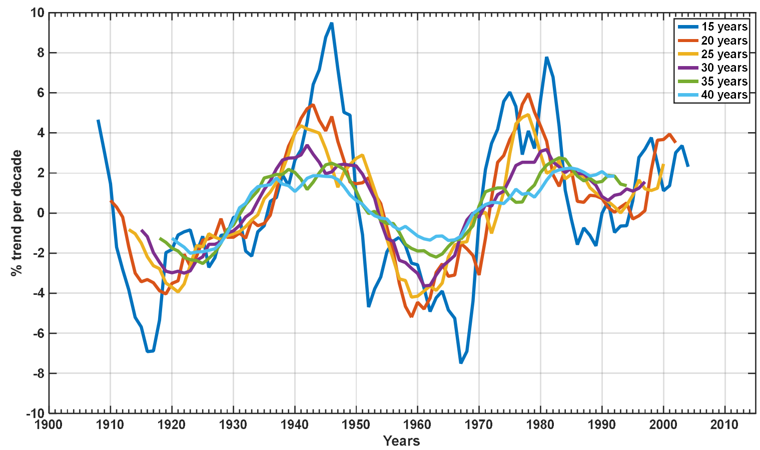
Table 2. Annual and seasonal SSR trends in percent per decade over the period 1900-2012 and different sub-periods. Percentages in parenthesis show the limits of the 95% confidence bounds.

Season	1900-2012	1900-1952	1953-1982	1983-2012
Winter	-0.15 (±0.46)	-0.68 (±1.26)	-6.43 (±3.83)	+0.52 (±3.26)
Spring	+1.05 (±0.38)	+0.15 (±1.08)	-0.60 (±3.10)	+2.77 (±3.10)
Summer	+0.54 (±0.31)	+0.43 (±0.78)	-1.14 (±2.90)	+1.38 (±2.55)
Autumn	+0.14 (±0.34)	+0.14 (±1.02)	-1.28 (±3.42)	-1.50 (±1.83)
Year	**+0.40** (±0.26)	**+0.02** (±0.73)	**-2.33** (±2.28)	**+0.80** (±1.96)

Season	1900-2012	1900-1952	1953-1982	1983-2012
Winter	-0.11 (±0.47)	-0.90 (±1.46)	-6.43 (±3.83)	+0.52 (±3.26)
Spring	+0.54 (±0.37)	+0.38 (±1.12)	-0.60 (±3.10)	+2.77 (±3.10)
Summer	+0.59 (±0.21)	+0.28 (±0.48)	-1.14 (±2.90)	+1.38 (±2.55)
Autumn	+0.21 (±0.44)	+0.11 (±0.97)	-1.28 (±3.42)	-1.50 (±1.83)
Year	**+0.39** (±0.22)	**+0.04** (±0.71)	**-2.33** (±2.28)	**+0.80** (±1.96)

Looking at the 1900-2012, period the seasonal and annual linear trends in SSR are less than 1% per decade. A positive change of 0.39% per decade has been calculated from annual values. For the whole data set, all seasons show positive trends, except for winter. For the periods with simulated SSR values (1900-1952), even smaller trends have been detected for spring and summer. The measuring period of 1954-2012 has been split into two sub-periods of 1954-1982 and 1983-2012. The first sub-period shows a negative annual change of -2.33% per decade in SSR, which is also reflected in all seasons with predominant changes during winter (-6.43% per decade). The second sub-period shows a positive trend of +0.80% per decade with highest ones in spring (+2.77% per decade) and summer (+1.38% per decade) and negative in autumn (-1.50% per decade). Looking at the trend significance described by the 95% confidence bounds, we can see significant positive trends –for 1900-2012 (yearly, summer and spring) and significant negative trends for yearly analysis and winter of 1953-1982.

In order to have a better understanding of the SSR changes over the 113-year period (1900-2012), we have calculated the decadal SSR trends for different time-windows (15 to 40 years). Figure 5 shows the results of this analysis.



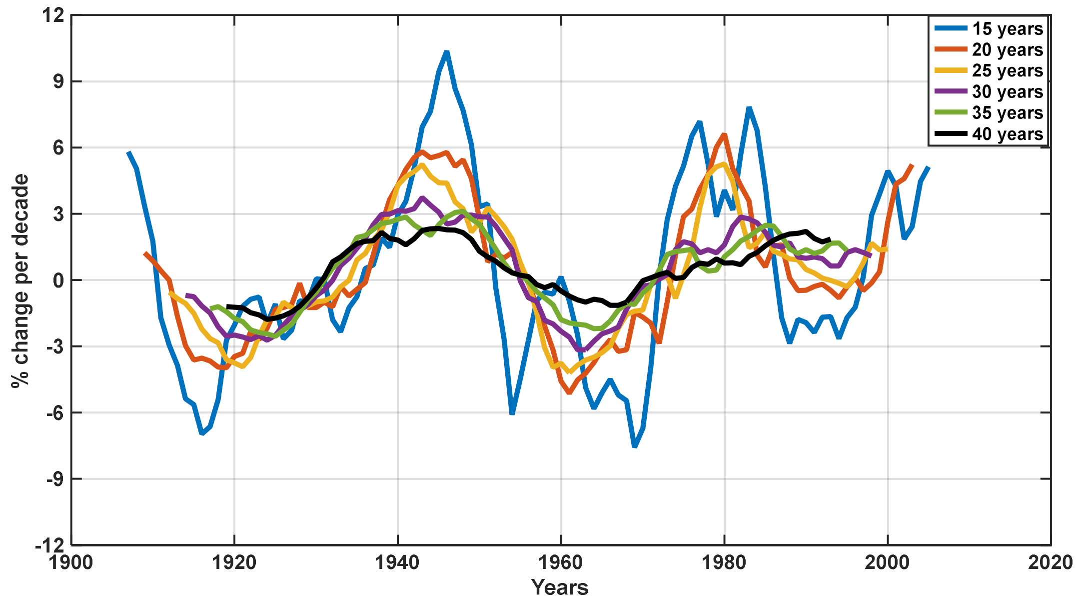


Figure 45. Trends in SSR (% per decade) calculated for different sliding time windows. The value of the trend has been calculated at the central year of each time window.

For the first two decades of the 20th century there appears a decrease in SSR, in line with other long-term SD series as recently shown by Stanhill and Achiman (2016). Then, in all calculations an increase is shown from mid 1930's to late 1940's, in line with the early brightening effect pointed out by other authors (Ohmura, 2009; Sanchez-Lorenzo et al., 2008). It should be reminded that this period is based on estimations of SSR from SD measurements, which thus include additional measurement uncertainties. Nevertheless, both the early dimming and brightening periods have been reported in Stanhill and Achiman (2016). and this study seemThe results can be partly supported by to be in line with trends in anthropogenic black carbon (McConnell et al., 2007; Lamarque et al., 2010) and biomass-burning (Lamarque et al., 2010) emissions in Europepeaking in

~~the 1920's and then decreasing~~. The dimming period from 1950's to 1970's can be observed in all time windows with a brightening effect after late 1970s.

The 40-year and 30-year time windows in the analysis presented in Figure 5 show the maximum rate of increase in early 1940's (resulting in an increase of 2% per decade and 3% per decade, respectively). Then a maximum rate of decrease is observed in early-mid 1960's, followed by a positive rate of increase after 1990's. Shorter time windows (15 years) are also interesting as they are able to capture the Pinatubo effect in early 1990's.

3.2 Variations and trends in SSR for the 1954~~3~~-2012 measurement period

In order to further analyze the whole 59-yr SSR data set of this study, we have divided it in two 30-yr climatological sub-periods: 1954-1983, and 1983-2012 (the common year is meant to have equal (30 full year) duration for both periods). Investigating a possible seasonal dependence, the relative difference in SRR for every month from its mean monthly value of the whole measurement (1954~~3~~-2012) period was calculated.

Figure 5~~6,~~ shows the mean daily insolation for each month for the two sub-periods and the whole 59-year period. Examining the monthly average differences between the two periods, we observe that for spring and summer months these are of the order of 6%. In addition, for all months SSR differences of the 1983-2012 period compared to the 1954-1983 period are positive with an exception of November (-1.9%) and December (-1.2%). In general, the second measurement period shows a 3% to 8% larger monthly SSR than the first measurement period.

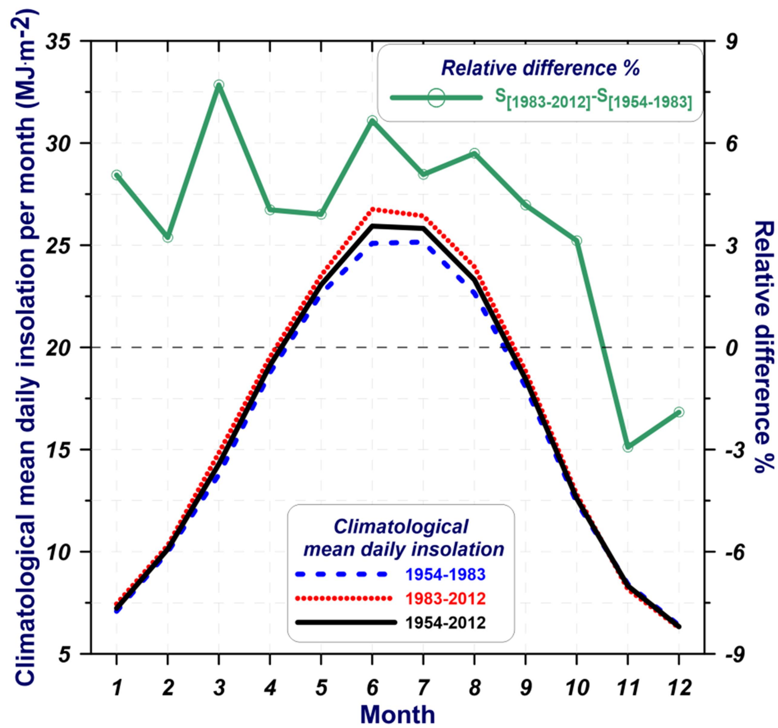


Figure 56. Intra-annual variability of monthly mean daily SSR over the sub-periods of 1954-1983 (blue line) and 1983-2012 (red line) and the entire period of 1954-2012 (black line). The green line (right axis) represents the monthly relative difference between the two 30-year sub-periods.

We have also calculated decadal trends in time windows of 15 to 30 years for the entire SSR measurement period (see Figure 7),. only for the 1954 to 2012 period, In the left panel of Figure 7 each of the points used for constructing the colored lines represents the percent change per decade of the SSR. Mostly positive trends are detected using any time window centered after 1975. Larger trends are calculated for time windows centered at 1975 to 1980 and after 2000 (in the order of 5% per decade using the 15-year time window). For the period 1954 to 1970 mainly negative trends are shown.

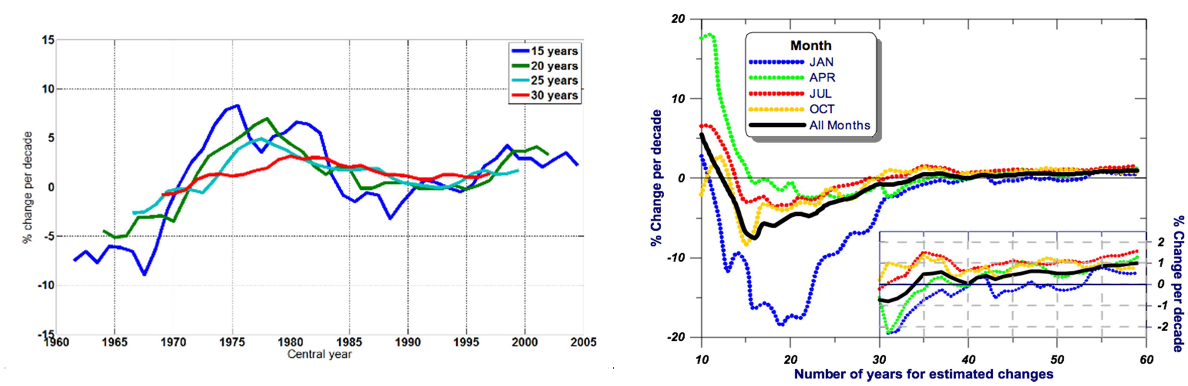


Figure 67. Left panel: calculation of SSR changes in % per decade for different time scales (15 to 30 years) used. Each point on the line represents the middle year of the time window used for the trend calculation Right panel: Per cent% change per decade for different time scales and different

months using 1954 as the starting year (the last 30 year period is magnified with changes presented as % per decade).

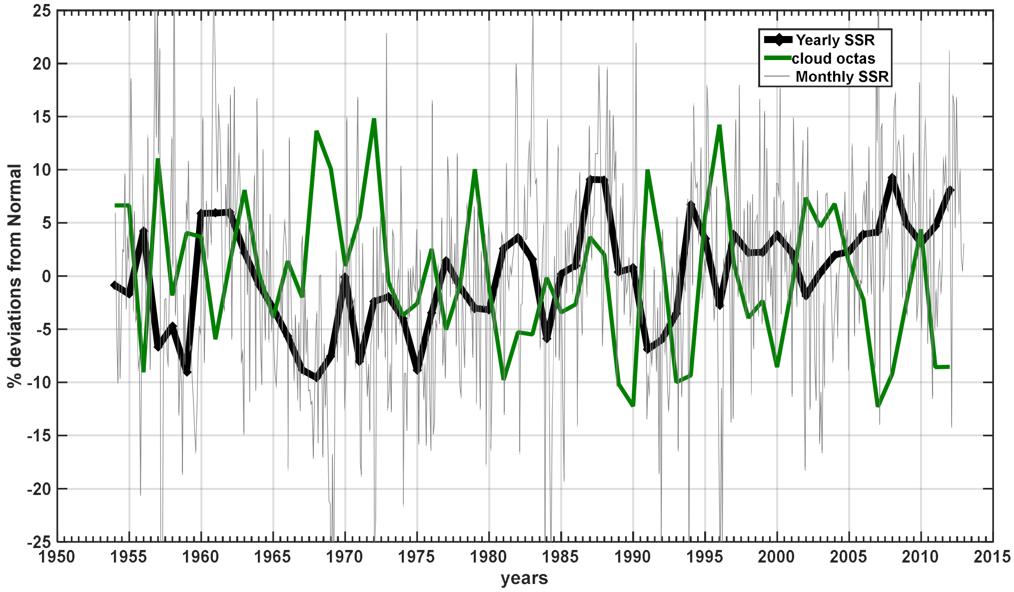
~~The right panel of~~ Figure 6~~7~~ shows the SSR change per decade for the months of January, April, July, October and yearly (all months). The figure is showing a trend analysis for the entire data set with time windows from 10 to 59 years, where each time window starts from 1953. For all months SSR changes become positive for time windows of 35 years and higher (1953-1988 time window and any larger window starting from 1953). Negative trends calculated from 1954 to any given year up to 1989 are mainly due to the large negative changes during the winter period. Especially during the 1954-1974 period, winter SSR changes show a 18% per decade decrease. Linear trends in SSR from 1954-2012 showed a positive trend of the order of 1% per decade, while individual months vary from 0.5% per decade to 1.5% per decade. Mostly positive trends are detected using any time window centered after 1975. Larger trends are calculated for time windows centered at 1975 to 1980 and after 2000 (in the order of 5% per decade using the 15-year time window). For the period 1954 to 1970 mainly negative trends are shown.

4. Comparison between all-sky and clear-sky SSR records variation

We have used the 59-year data set (1954~~3~~-2012) in order to quantify the factors controlling the SSR variations in Athens, Greece, focusing mainly on two known dominant factors, clouds and aerosol load.

4.1 The role of clouds

Figure 7~~8~~ shows the 1954-2012 time series of the monthly and yearly anomalies based on daily SSR, together with yearly total cloud coverage in weighted octas. The yearly de-seasonalised SSR values for all-sky conditions show a drop of ~14% from 1960 to 1970 and then a continuous increase excluding the Pinatubo period in the early 1990's. Most pronounced positive changes can be seen during the last 15 years with a change of the order of about 15%.



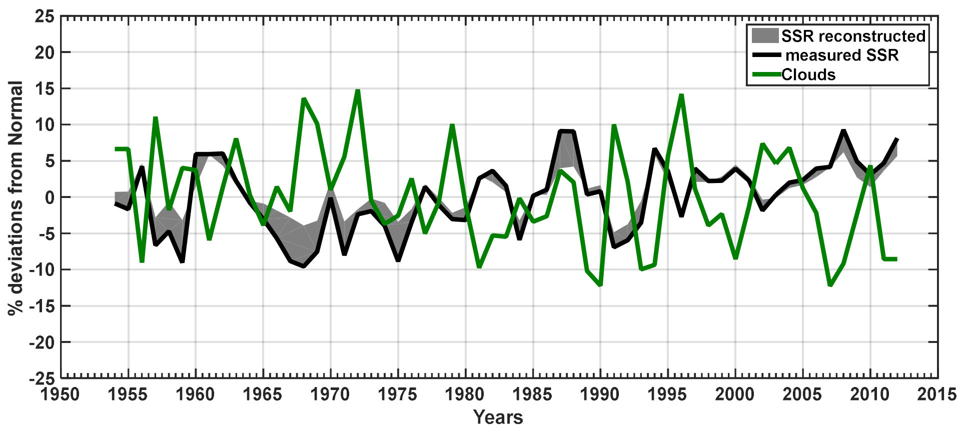


Figure 7~~8~~. De-seasonalised monthly mean (i.e. deviation from the respective monthly mean calculated over the whole period; dash line) and yearly mean SSR (black line).~~,~~ Grey lines are related with measurements possible uncertainties/drifts ~~along with mean annual total cloud cover (green line).~~

Figure 7~~8~~ suggests anti-correlation between variations in SSR and cloud cover.

Going back to the measurement uncertainties for the 1954-1983 period where a number of instruments have been used in order to build the presented time series; we have tried to investigate possible instrument drifts and their effect on the calculated long term trends. In order to indirectly try to tackle this issue we included in figure 7 a shaded area representing a possible (one direction) "uncertainty" based on reconstructing the 1954-1983 series using: the 1984-2012 measured SSR data and the sunshine duration data for 1954-1983. The reconstruction has been performed in the same way as the 1900-1953 one. The one direction “uncertainty”, points out possible drifts and instrument exchange related uncertainties. However, that does not mean that we believe more on the reconstructed through sunshine duration 1954-1983 series than the actual SSR measurements. If this was the case, we would have decided to present a 1983-2012 high quality measuring period and a 1900-1983 reconstructed one. There are various of such papers published quite recently (small measuring period compared with the reconstructed one: Garcia et al.,, 2014; [1992-2013 measurements reconstructed back to 1933] and Anton et al., 2017 [1887-1950 using radiative transfer modelling]) while in our case we would like to try to use the best way possible the historical SSR measurements of NOA during the 1954-83 period.

Using the 1984-2012 measurements and the 1900-1983 recinstruction data set we have recalculated all trends presented in figure 4 and table 2. Differences for the 15 year window differences on the calculated trends outside the 54-83 period are less than 1%, with maximum differences at the late 60’s 1-3%. For the 30 year window maximum differences are in the order of 1-2%., while for the 40 year window, maximum differences are less than 1%.

This particular exercise cannot be defined as an uncertainty assessment on the 1954-83 measurements, as reconstructed data cannot be used as a reference. Moreover, as SSR is much more sensitive than SD especially with respect to aerosol optical depth changes. So, in locations where the number of cloudless days is relatively high SD reconstruction tends to “smooth” the SSR variability, however the opposite can be said in cases with constant cloudiness.

Figure 89 shows the correlation between annual mean SSR and cloud cover. From the best-fit linear regression line it is seen that a -1.54 MJ m^{-2} (or -9.6%) change in mean daily insolation accounts for a change of 1 octa in cloud cover.

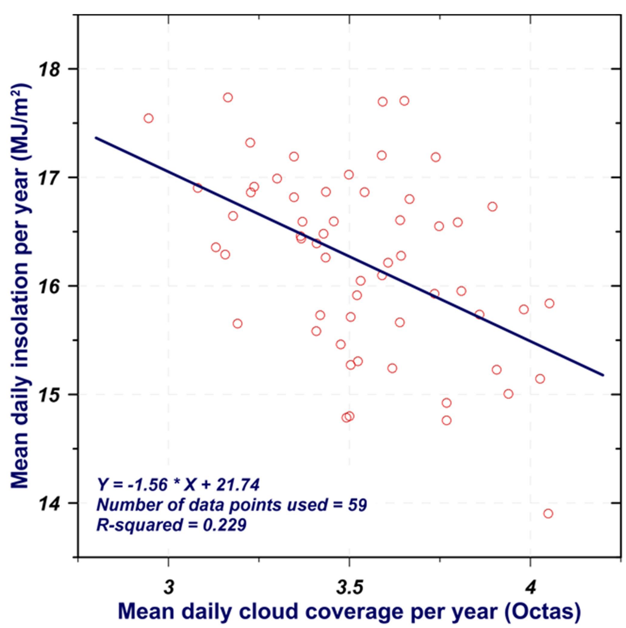


Figure 8~~9~~. Correlation between annual means of daily insolation and cloud cover over the period 1954-2012. The straight line represents the best-fit regression line to the data points. The year 1953 has not been included in the analysis since it does not include measurements for all months.

However, the great scatter of the data points and the low correlation of the two parameters in Figure 9 (R^2=0.229) indicate that the ~~presence of~~ cloud cover can only partly explain the changes in SSR. In addition, there is no significant change in cloudiness over the 59 year period for Athens, Greece. Calculating linear changes of cloudiness from data shown Figure 8, shows a ~~non~~ non-significant change of -0.4% per decade which can practically have a limited effect on SSR changes during the examined period.

Nevertheless, it is worth mentioning that different cloud properties like cloud optical thickness and cloud phase, not described by the measurements of cloud cover, can influence SSR.

4.2 Clear sky records

In order to minimize the cloud influence and investigate the possible role of direct aerosol effects on Athens SSR series, we had to select clear-sky (or cloudless) days. We have used daily SSR measurements from 1954~~3~~ to 2012 and we have separated the cloudless days according to the criteria mentioned in Section 2.2.

For considering the SSR seasonality, we have calculated a five-degree polynomial derived from the maximum daily SSR (for all years of the data set), as a function of the day of the year (Figure 9~~10~~). Afterwards we have calculated the ratio of the daily SSR to the SSR calculated by this function. Seasonal and yearly means of this ratio have been estimated and have been used to describe cloudless-sky SSR percentage changes on a seasonal and yearly basis. This approach has been chosen since averaging a random set of cloudless days, within each month during the 59-year period, could cause solar elevation-related (due to the change of maximum solar elevation within each month) discrepancies, when calculating the monthly average SSR. It can be emphasized that the clear sky selection criterion could possibly eliminate cases with very high aerosol optical depth.

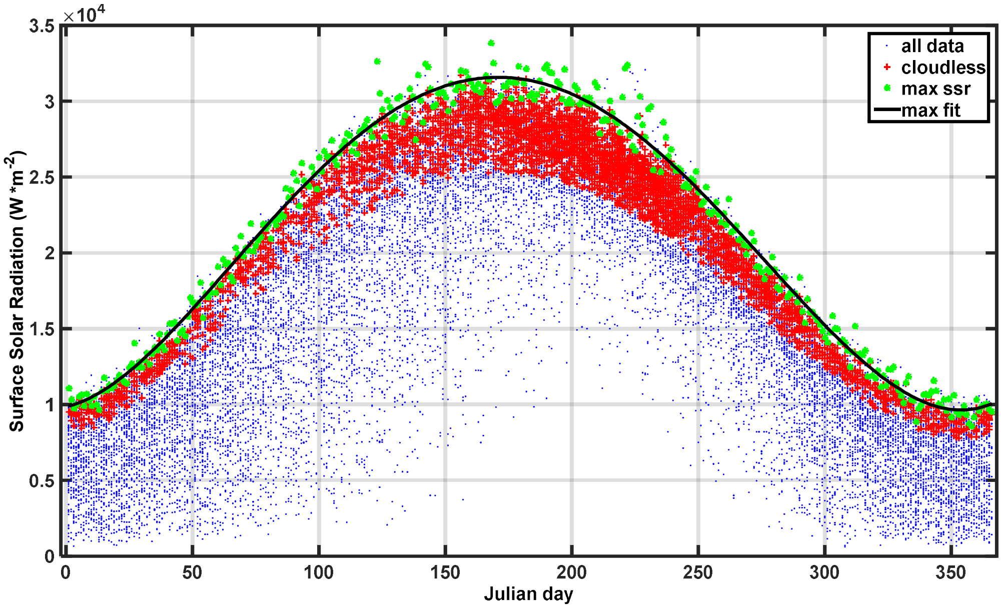


Figure ~~10~~9. Clear-sky SSR measurements (red dots) and all-sky SSR measurements (blue dots) derived with the cloud octa (cloudiness<1.5) and sunshine duration (SD>0.9) related criteria. The black line represents the polynomial fit to the daily SSR_{max} values.

Using the clear sky conditions seasonal and yearly averages of SSR have been calculated. The use of seasonal instead of monthly SSR has been introduced in order to improve the averaging SSR-related statistics, since the average number of cloudless days (per year) can be relatively low especially during the winter months. For all cases the ratios of the mean daily cloudless SSR to the SSR_{max} derived from the daily best-fit curve in Figure 10 has been calculated and deviations of this ratio from its 59-yr mean have been calculated for each year.

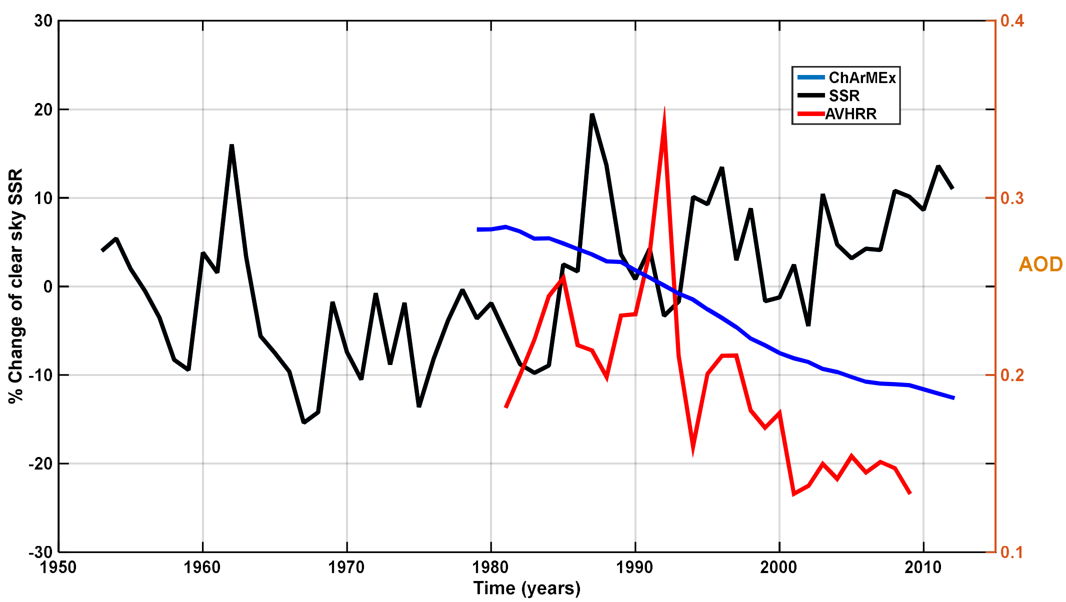


Figure 10~~1~~. Changes in yearly mean SSR to relative to the 1954-2012 average for cloudless sky (in %; ~~blue~~black), AVHRR AOD series (red) and ChArMEx AOD climatology (blue; Nabat et al., 2013) for Athens area is shown in the right axis.

Figure 10~~1~~ shows that most of the SSR variation observed for the measuring period has to be explained by other factors than changes in cloudiness (see figure 7~~8~~ for variations due to cloudiness). Different seasons with the exception of wintertime show similar patterns to the year-to-year variability. Individual seasonal calculated SSRs do not exceed by more than ±5%, the SSR variability of all sky data, with the exception of the winter season. Comparing clear sky and all sky yearly mean SSR, we find a high correlation (R^2 = 0.71), which can be explained as a combination of: aerosol changes driving the SSR changes and by the number of clear sky days during the year. There is a decrease of more than 15% in the clear sky SSR from the start of the series to the end of 1960's. A decline after 1983 could possibly be related with El Chichon volcanic eruption.

All sky SSR measurements and AOD from AVHRR have been used in order to find the AOD effect in all sky data. For yearly AOD and SSR averages from 1981 to 2009 a correlation coefficient of -0.55 was calculated with a rate of SSR reduction per 0.1 units of AOD equal with -3.8%.For monthly based comparisons, all months revealed a correlation coefficient of -0.2 with a rate of -1.5% per 0.1 AOD with better results for summer and Autumn months (-0.30, -2.2%/0.1 AOD and -0.30, -1.5%/0.1 AOD)

The Pinatubo-related drop of ~~5%~~6% from the early 1990's to the mid 1993, can also be seen in both cloudless and all-sky datasets and also to the increase in AOD in the AVHRR dataset (Figure 10~~1~~). Since, the ~6% drop from 1990 to 1991-1993 is shown for all seasons, we can argue that it describes the effect of the eruption on SSR data for the Athens station. However, as shown in figure 7 cloudiness for 1991 is also high, while is much lower for 1992 and 1993. Combined with the stratospheric AOD figure, seems that 1991 related decrease is also related with cloud increase while 92 and 93 one with the Pinatubo related aerosol effect.

Concerning the stratospheric AOD in Athens ChArMEx AOD dataset revealed two main peaks of 0.12 for 1983 and 0.09 for 1992 due to El Chichon and Pinatubo eruptions, respectively, while stratospheric AOD after 1995 is lower than 0.01. These two peaks are possibly associated with decreases in SSR as measured in ASNOA.

Finally, the ~13% change from 1995 to 2012 shown for all skies (Fig. 7~~8~~) and clear skies (Fig. 10~~1~~) is accompanied with a drop of ~25% in AOD measured by AVHRR. The year to year variations of clear sky SSR series and the AVHRR-related AOD show an anti-correlation with R=-0.78 (N=29), verifying the hypothesis that SSR clear sky changes are associated with aerosol load changes, at least within the common AVHRR/measurement period (1982-2009).

Similar to the AVHRR data the ChArMEx 4-D aerosol climatology is shown in figure 10~~1~~, providing similar conclusions, namely the AOD negative trend of 0.03 or 14% per decade from 1979 to 2012.

Differences between the AVHRR and ChaArMEx data can be explained in part by the different AOD wavelengths presented here (630 vs 550 nm) and also by a general negative bias of AVHRR over the Mediterranean compared to AERONET (Nabat et al., 2014). The smooth decline in the ChArMEx AOD data is due to the method used to build this product and uses the trend and not the interannual variability which is not included in the global model that was used.

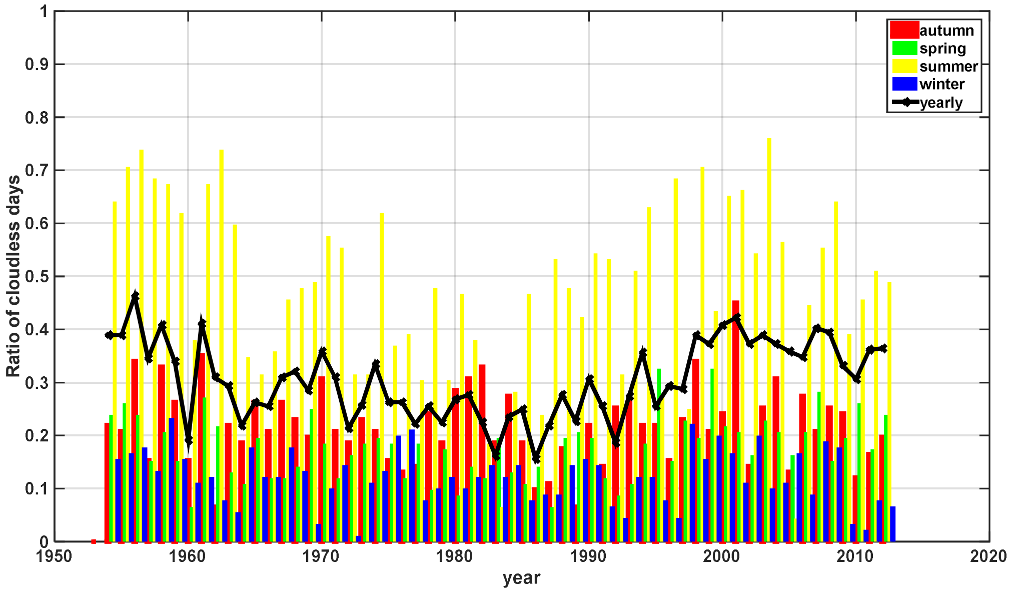


Figure 12. Ratio of cloudless vs all days, per season and yearly

In addition to figure 10~~1~~ we have included figure 11~~2~~ showing the ratio of cloudless days to all available days for each season and for each year. Figure 11~~2~~ shows a minimum (less than 30% during a year) of the number of cloudless days from mid-1970's to early 1990's. It is mostly linked with the decrease of cloudless days during summer months. The figure provides a hint on the SSR relative changes observed during this period, but it can not directly interpret year to year SSR changes as they depend also on cloud fraction and properties for cloudy days. In addition, it can only partly be linked with fig. 10~~1~~ as aerosol effects on cloudless sky calculated SSRs depend mostly on AOD levels and not on the number of days included in the calculations.

Differences on the ratio of cloudless days shown in figure 11 and on the almost constant cloud octa variability shown in figure 8 is partly attributed to the different definition of a cloudless day that is based on the cloud radiative effect for fig. 11 and on observation of cloud percentage in the sky for fig. 8. However, this can also be an indication of changes in cloud properties (e.g. change in optically thin clouds that could have small radiation effect but are marked as cloudy conditions from the observer).

In Table 3 we have calculated the linear trends for the 1953-2012 period and for both clear sky and all sky measurements and the 1953-1982 and 1983-2012 sub-periods for clear sky measurements. Results show comparable changes per decade (2% for the clear sky and 1.5% for the all sky cases). Seasonal analysis show that clear sky trends for summer, autumn and winter months are higher than the ones derived for all skies. Such differences are linked with the seasonal variability and long-term changes of cloudiness for the specific seasons.

Table 3: Clear sky and all sky data trends comparison for the whole 1953-2012 period and the two 30-yr sub-periods (% per decade). Percentages in parenthesis show the limits of the 95% confidence bounds.

Season	**Clear sky 1953-2012**	**All skies 1953-2012**	**Clear sky 1953-1982**	**Clear sky 1983-2012**
Winter	0.91 (±2.31)	-6.43 (±3.83)	-7.01 (±3.16)	0.55 (±2.41)
Spring	1.22 (±1.12)	-0.60 (±3.10)	-0.92 (±1.11)	2.62 (±1.97)
Summer	2.03 (±0.78)	-1.14 (±2.90)	-0.36(±0.83)	1.31(±0.81)
Autumn	2.74 (±1.37)	-1.28 (±3.42)	-1.03(±1.84)	-1.48 (±1.73)
Year	**2.17 (±1.21)**	**-2.33** (±2.28)	-1.44(±2.35)	1.94 (±2.08)

Clear sky results for the 1953-2012 period show significant positive changes in SSR for all seasons except winter. Looking individually at the 1953-1982 and 1983–2012 periods we have calculated significant negative trends only for the winter over the first and for summer and spring over the second.

The effect of various parameters on SSR has been discussed by Kambezidis et al. (2016) in their study about the global dimming/brightening effect over the Mediterranean in the period 1979-2012; they show that the influence of parameters related to the atmospheric transparency, like water vapor, aerosols and trace gases, as well as changes in the surface albedo on SSR have been larger in the southern parts of the Mediterranean, over the Balkan countries and central Turkey. This outcome is in agreement with the conclusion of the present study that other factors than cloudiness play significant role in the SSR variations.

A comparison of the SSR results in Athens with visibility observations since 1931 (Founda et al., 2016) did not show any correlation among SSR and horizontal visibility. For the first part of the common dataset (1930-1959) the visibility decline is accompanied with a SSR increase. However from 1950 till today visibility shows a monotonical decrease. The steep visibility decrease from 1931 till early 90's is not accompanied by a relative SSR decrease excluding individual sub-periods. However, simulated SSR is driven purely by changes in sunshine duration, in this case the SD variability in founda et al., 2014 is almost stable after 1950 so SD can not be also linked with the visibility reported decrease. Studying the literature for similar cases, similar conclusions have been drawn by Liepert and Kukla (1997) showing an SSR decrease over 30 years of measurements accompanied by a visibility increase and no significant changes in the cloud cover conditions, in Germany. This Athens SSR vs visibility relationship can be partly explained by the fact that: SSR and visibility have different response on cloud conditions, water vapor and rainfall and also by the fact that visibility is affected by aerosols only in the first few hundred meters above the surface, while SSR is affected by the columnar AOD, which in the case of Athens can be significantly different due to aerosol long-range transport in altitude (e.g. Saharan dust; Léon et al., 1999).

5 Conclusions

Surface solar radiation (SSR) at National Observatory of Athens, in the center of the city, is presented using a unique dataset covering a period of 59 years (1954-2012). Sunshine duration (SD) records for another 54 years have been used as a proxy to reconstruct SSR time series for the period from 1900 to 2012.

The data accuracy of such historic radiation dataset is more difficult to be assessed especially going back to the 50' and 60's where instruments, operational procedures and quality control were not in the same level as in the recent 30 years. Quality assessment procedures in the presented time series have been applied with criteria based on instrument characteristics and the availability of additional collocated measurements. Year to year fluctuations of the measured SSR in addition to the reversal of the downward tendencies at the ASNOA site adds credibility to the measured variations. That is because a typical radiometer behavior is to lose sensitivity with time indicating spurious downward, but not upward trends. The more recent (after 1986) SSR measurements can be characterized as high-quality radiation data with known accuracy. Considering the measurements from 1954 to 1970 there has been sporadic reports mentioning the homogenization and calibration procedures, while for 1970 to 1986 there is more information on the instrument quality control.

Reporting of the results from the 1954-1986 period should be accompanied with the fact that the uncertainties of the measurements of this period are linked with higher uncertainties than after 1986. For the reconstruction of the 1900-1953 series, only the 1983-2012 SSR and SD measurements used in order not to link possible instrument uncertainties to the extrapolated period.

However, reconstruction the 1900-1983 time series using the 1984-2012 dataset leads to small differences in the determination of the long term trends, especially for more than 20 year running average windows.

A comparison of the SSR results in Athens with visibility observations since 1931 (Founda et al., 2016b) did not show any correlation among SSR and horizontal visibility. The steep visibility decrease from 1931 till early 90's is not accompanied by a relative SSR decrease excluding individual sub-periods. Studying the literature for similar cases, similar conclusions have been draown by Liepert and Kukla (1997) showing an SSR decrease over 30 years of measurements accompanied by a visibility increase and no significant changes in the cloud cover conditions, in Germany. This Athens SSR vs visibility relationship can be partly explained by the fact that: SSR and visibility have different response on cloud conditions, water vapor and rainfall and also by the fact that visibility is affected by aerosols only in the first few hundred meters above the surface, while SSR is affected by the columnar AOD, which in the case of Athens can be significantly different due to aerosol long-range transport in altitude (e.g. Saharan dust; Léon et al., 1999). The data accuracy of such historic radiation measurements is generally not well established, at least compared with the measurements after the 1990s. Quality assessment procedures in the presented time series have been applied based on criteria based on instrument characteristics and the availability of additional collocated measurements. Year to year fluctuations of the measured SSR in addition to the reversal of the downward tendencies at the Athens site adds credibility to the measured variations. That is because a typical radiometer behavior is to lose sensitivity with time indicating spurious downward, but not upward trends. The more recent (after 1989) SSR measurements can be characterized as high-quality radiation data with known accuracy.

De-seasonalized SSR data analysis from 1900 to 2012 showed high month to month variability that could reach up to 25%, mainly related with monthly cloudiness variations. During the period 1910-mid-1930s where only few datasets have reported worldwide SSR results, we observe a 2.9% per decade or a total of 8.7 % decrease in SSR, assuming linear changes in SSR during this period. This early dimming was followed by a 5% per decade increase from 1930 to the 1950s. Similar results have been found at Washington DC and at Potsdam, Germany (Stanhill and Achiman, 2016). They have reported an early brightening at both locations in the 1930's. For the SSR measurement period of 1953 to 1980, European related studies presented in Wild (2009) showed a -1% down to -7% change per decade in SSR measurements over various European sites (dimming period). For the Mediterranean region, Manara et al. (2016) showed a decrease in the order of -2% to -4% per decade in Italy. We are reporting a change in SSR of -2% per decade in Athens. Finally, for the brightening (1990-2012) phase again Wild et al. (2009) reported a 1.6% up to 4.7% per decade positive change in SSR while we have calculated a +1.5% per decade, which is lower than the 3-6% per decade reported in Manara et al. (2016) for Italy. A summary of the above findings can be seen in table 4.

Table 4: Summary of per cent SSR changes per decade for various locations

Period	**Location**	**Trend % per decade**	**Reference**
1893-2012	Potsdam, Germany	0.71	Stanhill and Achiman, 2014
1900-2012	Athens, Greece	0.40 (±0.26)	This work
1959-1988	Europe	–2.0	Ohmura and Lang, 1989
1971-1986	Europe	–2.3	Norris and Wild, 2007
1959-1985	Italy	–6.4(±1.1) / -4.4(±0.8)	Manara et al, 2016
1953-1982	Athens, Greece	-2.33(±2.28)	This work
1985-2005	Europe	2.5	Wild, 2009
1990-2012	Italy	6.0 (±1.1) / 7.7 (±1.1)	Manara et al, 2016
1986-2013	Athens, Greece	0.80 (±1.96)	This work

The decadal variations of SSR measured since 1954 at Athens, Greece, originate from the alterations in the atmosphere's transparency (namely by clouds and aerosols). Using an analysis of SSR calculations of all sky and clear sky (cloudless) days we end up that since cloud cover changes during the 59 period were very small, most of the observed decadal changes might be related with changes in the aerosol load of the area. An additional hint in support of this conclusion is the high correlation of clear sky and all sky yearly SSR. We also found an anti-correlation between either clear sky and all sky SSR measurements and AOD time series from AVHRR (1981-2009) or

ChArMex (1979-2012). Looking at linear trends over the 59 year period clear sky changes per decade were 2% while it was 1.5% for all sky conditions. The most pronounced changes have been calculated for summer and autumn seasons (2% and 2.7% respectively).

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