

Decadal variability of aerosol optical depth in Europe and its relationship to the temporal shift of the North Atlantic Oscillation in the realm of dimming and brightening

Marc Chiacchio,¹ Tracy Ewen,^{1,2} Martin Wild,¹ Mian Chin,³ and Thomas Diehl³

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[1] Long-term aerosol optical depth (AOD) over Europe was analyzed from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model from 1979–2007. In particular, we studied the decadal sulfate AOD variability caused by large sulfur emissions from anthropogenic sources in Europe with a peak in 1988–1989. Simulated annual means from 1985–2007 over the continent showed statistically significant declines of 69% and a maximum of 75% in eastern Europe. Seasonally, greatest variations occurred during winter and spring followed by summer and autumn. The decrease in AOD agrees with the increase in the annual, spring, and summer mean solar radiation in Europe after the mid-1980s as well as surface-based AOD measurements. However, the long-term AOD variability does not explain the trends found in solar radiation during winter and autumn, which may be due to the contribution from the North Atlantic Oscillation (NAO) and associated cloud cover. We also investigated a possible link between sulfate AOD and NAO and found a statistically significant correlation of -0.77 in winter for Europe. Wavelet coherence analysis revealed a strong and significant antiphase relationship around 1 year that was most pronounced in the late 1980s. Cross-correlation analysis showed a seasonal dependence of sulfate AOD and NAO with negative correlation during winter and positive during summer. This analysis may help explain the seasonal decadal variability of surface solar radiation and whether sulfate aerosols have contributed to the large positive trend of the NAO during the 1980s. However, the cause and effect relationship between sulfate aerosols and NAO remains unclear.

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1. Introduction

[2] One of the largest uncertainties in climate change research today is the computation of the radiative forcing of aerosols on climate [Forster *et al.*, 2007]. This is mainly due to the aerosols' short lifetime in the atmosphere (about 1 week), which makes it difficult to study their temporal and spatial global distribution and their physical and chemical properties. Their treatment in most global and regional climate models is now being considered, but complete understanding of their effects on the climate is still not well-known. Depending on the type of aerosol, they can either scatter or absorb solar radiation [Tegen *et al.*, 2000; Novakov *et al.*, 2003], and through indirect effects they can alter cloud microphysical properties [Lohmann and Feichter,

2005]. These aerosol-radiative processes are known to either lead to an enhancement or a decline in surface temperatures over bodies of water [Lelieveld *et al.*, 2002; Evan *et al.*, 2009], which can affect atmospheric circulation [Rodwell *et al.*, 1999; Bojariu and Gimeno, 2003; Paeth *et al.*, 2003; Kucharski *et al.*, 2006; Sun *et al.*, 2009] and possibly the Northern Hemispheric climate [Luo *et al.*, 2009]. Hence, the knowledge of their distribution and microphysical properties, their physical mechanisms, impacts, and long-term variability, especially for tropospheric anthropogenic sulfate aerosols, is essential for model development and for studying the climate.

[3] During the past couple of decades the emissions of sulfate aerosols in Europe have declined [Vestreng *et al.*, 2007; Berglen *et al.*, 2007], after a maximum of emissions was reached in 1988–1989 [Streets *et al.*, 2006], due to environmental regulations imposed to control air pollution, which also resulted in an increase in visibility [Vautard *et al.*, 2009; Wang *et al.*, 2009]. This decline in emissions of sulfate aerosols is also in line with previous studies made with annual mean global and regional surface and satellite observations of total aerosol optical depth [Mishchenko and Geogdzhayev,

¹Institute for Atmospheric and Climate Science, ETH Zurich, Zurich, Switzerland.

²Also at Department of Geography, University of Zurich, Zurich, Switzerland.

³NASA Goddard Space Flight Center, Greenbelt, Maryland, USA.

2007; Ruckstuhl *et al.*, 2008; N. Hatzianastassiou *et al.*, manuscript in preparation, 2010] and decadal variations of satellite-derived aerosol optical depth in general [Torres *et al.*, 2002]. With decadal changes in the emission of aerosols, it is believed they have influenced the amount of solar radiation that reaches the surface of the Earth, upsetting the surface radiative balance, hence, global dimming and brightening [Ohmura and Lang, 1989; Gilgen *et al.*, 1998; Stanhill and Cohen, 2001; Liepert, 2002; Wild *et al.*, 2005; Pinker *et al.*, 2005; Wild, 2009] and possibly contributing to the temperature rise in Europe since 1980 [Ruckstuhl *et al.*, 2008]. This phenomenon also has an impact on the diurnal temperature range and sunshine duration [Makowski *et al.*, 2009; Sanchez-Lorenzo *et al.*, 2008]. Though it has been shown that clouds also play a role in decadal changes in surface solar radiation [e.g., Dutton *et al.*, 2006], the interrelationships between these variables are still not well understood and they have not yet been quantified. Studies have also focused on regional and finer temporal resolutions [Sanchez-Lorenzo *et al.*, 2008; Stjern *et al.*, 2009; Chiacchio and Wild, 2010; Chiacchio *et al.*, 2010], such as the seasons in Europe, and have found that during winter and autumn the North Atlantic Oscillation (NAO), through a modification of cloudiness, may contribute to the decadal changes in the all-sky solar radiation reaching the surface [Chiacchio and Wild, 2010]. Moreover, it was proposed that during these two seasons aerosols could be interfering and strengthening the relationship between solar radiation and the NAO.

[4] It is well established that the NAO mainly affects the climate in Europe in winter [Hurrell and van Loon, 1997; Bojariu and Giorgi, 2005] and is a dominant pattern of climate variability in the North Atlantic region [Hurrell *et al.*, 2003]. It is also one of the teleconnection modes that persist throughout the year including the summer [Hurrell *et al.*, 2003; Folland *et al.*, 2009; Sun *et al.*, 2009]. However, the mechanisms that involve its changes through time are not yet fully understood [Bojariu and Gimeno, 2003; Gillett *et al.*, 2003], and its role in global dimming and brightening is only now being investigated. In recent decades, large changes in the NAO have occurred with a large positive trend in the index in the 1980s, and it has accounted for much of the warming in Europe [Hurrell and van Loon, 1997; Trigo *et al.*, 2002; Gillett *et al.*, 2003]. Moreover, it has been suggested that the upward rise in the NAO index might be the result of anthropogenic forcing on the climate [Visbeck *et al.*, 2001; Bojariu and Gimeno, 2003] with a focus on the role of greenhouse gas forcing on the NAO [Osborn *et al.*, 1999; Ulbrich and Christoph, 1999; Paeth *et al.*, 1999; Osborn, 2004; Coppola *et al.*, 2005; Stephenson *et al.*, 2006]. Though some models show an upward trend in the NAO when they are forced with increasing greenhouse gases, the magnitudes of these changes is inconsistent with the real atmosphere and not all models reproduce these changes [Gillett *et al.*, 2003]. It has been shown that the NAO influences dust transport, for example, from North Africa into the Mediterranean [Moulin *et al.*, 1997; Antoine and Nobileau, 2006; Dayan *et al.*, 2008], and also it has been indicated that variations in African dust aerosols over the subtropical Atlantic Ocean, through their impact on the sea surface temperatures, may be a physical mechanism for the phase change of the NAO [Luo *et al.*, 2009]. In addition, recent studies have investigated anthropogenic aerosol forcing on

atmospheric dynamics [e.g., Kim *et al.*, 2007; Zanis, 2009; Allen and Sherwood, 2010] and in particular the effect of sulfate aerosols on planetary stationary waves and their dynamic feedbacks on temperature, precipitation, and surface pressure patterns resembling the NAO [e.g., Lewinschal and Ekman, 2010]. A study by Fischer-Brunns *et al.* [2009] focuses on a future simulation of the impact of anthropogenic sulfate aerosol on the North Atlantic winter climate and finds an anticorrelation with NAO suggesting that it could be affected by changes in this type of aerosol.

[5] It is inconclusive whether tropospheric anthropogenic sulfate aerosols have an influence on the NAO. However, because the large positive trend in the NAO also occurred at about the same time when aerosol emissions in Europe were at a maximum, it might imply that this type of aerosol, through, for example, the modification of sea surface temperatures (SSTs), could be contributing to the changes in the NAO. As a consequence, this may affect the decadal variation in surface solar radiation differently according to the season as found by Chiacchio and Wild [2010], where the winter trend during 1985–2000 was found to be near zero and was mainly influenced by the NAO and its associated cloud cover. The trends of the surface solar radiation in spring and summer, however, showed an increase during this period. This is the motivation for our study. We first construct an annual and seasonal time series of sulfate and total aerosol optical depth (AOD) over Europe and its subregions simulated by the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model for the period 1979–2007 and then detect trends for 1985–2007 in order to be consistent with the beginning of the brightening period considered by Chiacchio and Wild [2010]. The performance of the model will be assessed by comparing annual mean long-term variations of total AOD to surface measurements of the same aerosol type from six stations given by Ruckstuhl *et al.* [2008]. This validation might also show whether these point measurements can be spatially representative for grid scale changes in AOD. Seasonal AOD trends will possibly help to further explain the same temporal scale decadal variability of solar radiation during dimming and brightening. Finally, an attempt is made for the first time to elucidate a possible link from mainly a wavelet-based statistical approach between sulfate AOD and the NAO and whether this analysis sheds more light on the mechanism that might be partially responsible for the large positive NAO index during the 1980s.

2. Data and Methods

2.1. Goddard Chemistry Aerosol Radiation and Transport Model

[6] The sulfate AOD used in this study is from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model, which has a horizontal resolution of 2° latitude by 2.5° longitude with 20–55 vertical sigma layers [Chin *et al.*, 2002]. Assimilated meteorological fields used as input are constrained by meteorological observations and are generated from the Goddard Earth Observing System Data Assimilation System (GEOSDAS). The most recent sulfur, dust, and biomass burning emissions are incorporated into the model. Anthropogenic sulfur emissions are from the Emission Database for Global Atmosphere Research (EDGAR)

[Chin et al., 2002; Olivier et al., 1996]. Chin et al. [2002] provides a more detailed description for the source of each type of aerosol. The model simulates a global distribution of AOD from various aerosol types, such as sulfate, dust, sea salt, and carbonaceous (organic and black carbon). Total AOD from this model is the sum computed from each aerosol type. The calculation of the AOD used in this model is performed at a wavelength of 500 nm and is determined from the simulated aerosol mass and optical properties from the Global Aerosol Data Set (GADS) [Kopke et al., 1997; Chin et al., 2002]. For the performance of the GOCART model against sun photometer measurements, please see the article by Chin et al. [2002].

[7] The advantage of using models for a long-term study of AOD is their great spatial and temporal coverage compared to the AOD provided by surface measurements and satellites which do not provide such extensive coverage. In order to assess the long-term changes in AOD from the simulations and the overall representativeness of site measurements, we show a comparison of changes in total AOD from six sites in Switzerland and Germany (Zingst, Lindenberg, Payerne, Hohen Peissenberg, Davos, and Jungfrauoch) of surface-based measurements given by Ruckstuhl et al. [2008] with the same collocated grid points of total AOD at 500 nm in the GOCART model and compute the trend during their common period 1986–2005. To avoid any potential influence from the Mount Pinatubo volcanic eruption in the model, we do not take into account the years 1991–1997.

2.2. Estimation of Sulfate and Total AOD Trends

[8] Sulfate and total AOD annual and seasonal mean trends from this model for the period 1985–2007 were also estimated for all of Europe as well as for different subregions, such as the northern, southern, eastern, and western parts. The year 1985 was chosen in the trend analysis in order to be consistent with the start of the brightening phase given by Chiacchio and Wild [2010]. A linear regression model of the first order was used to determine the best fit line. Confidence intervals at the 95% level were computed to test the statistical significance of the trends using the Student's t test with a p value equal to 0.05 and the standard error of the fitted line. If the null hypothesis is rejected or when the trend line is different from zero, then the trend is statistically significant at the 95% confidence level. The autocorrelation of the time series for different time lags was also considered, which can detect randomness in a time series and support the test for significance. A time series is random when the autocorrelations in all time lags are within the 95% confidence limits or are within $\pm(2/N^{1/2})$, where N is the number of data points in the time series. All time series analyzed for the estimation of the trends were found to be random.

2.3. North Atlantic Oscillation Index

[9] The seasonal values of the NAO index used for the correlation analysis with the sulfate AOD were taken from the results of Jim Hurrell of the Global and Climate Dynamics Division (CGD) Climate Analysis Section at the National Center for Atmospheric Research (NCAR) in Boulder, Colorado, USA (<http://www.cgd.ucar.edu/cas/jhurrell/indices.html>). This index represents the principal component (PC) time series of seasonal sea level pressure (SLP) anomalies

over the Atlantic region (20°N–80°N, 90°W–40°E). Monthly values of the NAO index were calculated from the Met Office Hadley Centre's mean sea level pressure data set (HadSLP2) fields [Allan and Ansell, 2006], based on the pointwise method. This was achieved by calculating the difference in normalized mean sea level pressure from the HadSLP2 data set with a horizontal resolution of $5^\circ \times 5^\circ$ between Ponta Delgada, Azores, and Reykjavik, Iceland.

2.4. Correlation of Sulfate AOD and NAO

[10] Statistical techniques used to study the relationship between the sulfate AOD and NAO were based on the computation of the Pearson correlation coefficient. Statistical significance at the 95% confidence level of this coefficient was determined from the critical t value from the Student's t test with a p value equal to 0.05, which was compared to a t value from $t = r[(n - 2)/(1 - r^2)]^{1/2}$, where n is the number of data points and r is the correlation coefficient. If the null hypothesis was rejected, then the correlation was statistically significant. The correlation coefficients were computed for the periods 1979–1990 and 1998–2007, for all seasons, i.e., winter (December, January, and February (DJF)), spring (March, April, and May (MAM)), summer (June, July, and August (JJA)), and autumn (September, October, and November (SON)), and for Europe and the subregions as mentioned earlier. The sulfate AOD and NAO time series were detrended by removing or subtracting the estimated linear trend from the original series. A moving average of 5 years was used in order to smooth the time series to study the correlation in the low-frequency variability. Spatial correlation maps for every season for sulfate AOD and NAO were also calculated for both periods. The annual cycle of the cross correlation between these two variables was analyzed for the two periods and for all subregions. Statistical significance at the 95% confidence level was determined as above.

2.5. Wavelet Analysis

[11] In order to examine variations of power through time within a time series, it becomes necessary to use wavelet analysis. This technique decomposes a time series into time-frequency space to determine the dominant modes of variability and how they change through time (nonstationarities) as opposed to transformation into only the frequency dimension, which other more classical methods use including Fourier analysis. The approach used here for applications to geophysical data is based on that of Torrence and Compo [1998]. In particular, the continuous wavelet transform (CWT) was applied to determine if a time series shows a nonstationary behavior at different frequencies or periods. The nonstationarity of the NAO has been studied before using wavelet analysis [Appenzeller et al., 1998; Loboda et al., 2006; Massei et al., 2007], but here we applied the method to monthly instead of annual means, which reveals the NAO's dominant mode of variability on a monthly time scale. Moreover, we also applied the CWT to the sulfate AOD for the period 1979–2008. We analyzed both time series before and after the Pinatubo eruption due to the slow decay of the simulated AOD, by removing the period 1991–1997. Statistical significance at the 95% confidence level for both CWT series was estimated against a red noise background [Torrence and Compo, 1998].

Table 1. Change in Total Aerosol Optical Depth (AOD) From Station Measurements Taken From *Ruckstuhl et al.* [2008] and Colocated Box Values Derived From the Goddard Chemistry Aerosol Radiation and Transport (GOCART) Model Over the Periods 1986–2005 and 1998–2005^a

Site (Location)	Total AOD Change (%)	
	SFC Measurement (1986–2005)/GOCART (1986–2005)	SFC Measurement (1995–2005)/GOCART (1998–2005)
ZIN	-63/-42	-27/-28
LIN	-60/-47	-21/-22
PAY		-26/-2
HOP		-15/-10
DAV		-12/-6
JUN		-10/-5

^aStation abbreviations: ZIN, Zingst; LIN, Lindenberg; PAY, Payerne; HOP, Hohen Peissenberg; DAV, Davos; JUN, Jungfrauoch; SFC, surface. Bold values indicate statistical significance at the 95% confidence level.

[12] In order to examine the link between the two series and whether their common power has a consistent phase relationship, we used wavelet coherence [*Grinsted et al.*, 2004]. It is proposed that if the phase relationship is indeed consistent, then their power is phase locked, i.e., arrows are pointed in only one direction for each significant power, which might suggest a cause and effect from the two series. Monte Carlo methods with red noise were used to determine the 95% statistical confidence level.

3. Results

3.1. Analysis of Long-Term Sulfate AOD

[13] The long-term change of total AOD from the GOCART model was compared to the change of total AOD from six sites in Switzerland and Germany (Table 1) given by *Ruckstuhl et al.* [2008]. Overall there was a decrease found in the total AOD from GOCART, especially for the Zingst and Lindenberg colocated sites with an average statistically significant decline of about 44% for the period 1986–2005. A slightly larger average statistically significant reduction of about 62% from the measurements of these two sites was observed, but not the same temporal coverage is used in the analysis. *Ruckstuhl et al.* [2008] removed the years 1991–1994 due to the eruption of Mount Pinatubo. For the period 1998–2005, both modeled results for the colocated sites for Zingst and Lindenberg compared well with the measurements. An average decline of about 25% from the model was found compared to an average of 24% from the measurements and were all found to be statistically significant. If only the magnitude of the trend was considered from these two locations, then the model results were nearly identical to the site measurements for 1986–2005 but were larger by about a factor of 2 for the period 1998–2005.

[14] Additional results of GOCART from other colocated sites as shown in Table 1 (Payerne, Hohen Peissenberg, Davos, and Jungfrauoch) for the period 1998–2005 also displayed a decline in total AOD with an average of about 6%, but none of them were statistically significant. Even though the model underestimated the percent changes of these site measurements, their actual trends for Hohen Peissenberg, Davos, and Jungfrauoch when compared to the site mea-

surements were greater than by a factor of 2 except for Payerne. The modeled result for Payerne displayed the greatest discrepancy when compared to the measurements.

[15] Figure 1 depicts the annual and seasonal time series for the period 1979–2007 for sulfate AOD in Europe. Sulfate and total AOD trends for the period 1985–2007 were estimated from the best fit line and were all found to be statistically significant at the 95% confidence level. The seasonal mean series mirror the behavior of the annual mean series, but differences among their trends are apparent. This was also true for the comparison between the annual and seasonal mean series for the total AOD (figure not shown). Among the seasons for the sulfate AOD for Europe, we found spring to display the largest negative trend followed by winter, summer, and autumn. The largest decline for total AOD was shown in winter, then in spring, summer, and autumn. Also, all sulfate trends are only slightly smaller than those from the total AOD. Table 2 shows that the estimate for the annual change in sulfate (total) AOD for Europe was -69% (-41%), while northern, southern, eastern, and western Europe had a decline between a minimum of 63% in southern Europe (33% in western Europe) and a maximum of 75% in eastern Europe (50% in eastern Europe). The seasonal mean changes for the sulfate (total) AOD for these subregions ranged between -48% in summer for western Europe (-26% in summer for western Europe) and -132% in winter for northern Europe (-77% in winter for northern Europe), and all were statistically significant.

3.2. Sulfate AOD Relationship with NAO

[16] Correlation analysis was performed on all regional and seasonal time series between sulfate AOD from GOCART and the NAO index for the periods 1979–1990 and 1998–2007. Their correlation for the period 1979–1990 in the detrended and low-frequency variability series (5 year moving average) was good, especially in the winter and autumn, and all were statistically significant, except for summer. In winter values of -0.77 and -0.76 were observed for Europe and averaged from all regions, respectively. In spring there was a slight decrease in correlation from an average of -0.51 to an average of -0.42 when compared to the high-frequency variability or unfiltered time series. The 5 year filtered series in summer has a higher correlation coefficient than its raw time series (high frequency) in all regions, but the average was only 0.25. An average of 0.55 from all regions was found in autumn. Overall for the period 1979–1990 in all regions during the winter and spring, the correlation is negative and is statistically significant with good to moderate values. The summer, on the other hand, had low and nonsignificant positive values. Finally, there is a fair and positive correlation in autumn and it is statistically significant with an average of 0.55. The correlation for the period 1998–2007 ranges from low to good values for the different regions and seasons with a maximum of -0.69 during spring in eastern Europe and is statistically significant in the high-frequency variability series. Also, there is not a consistent pattern of positive and negative correlation for each season.

[17] Spatial correlation maps for the periods 1979–1990 and 1998–2007 between the sulfate AOD and NAO were also constructed which showed a clear dipole pattern of statistically significant negative correlation in the central to the northern part of Europe and positive significant correlation

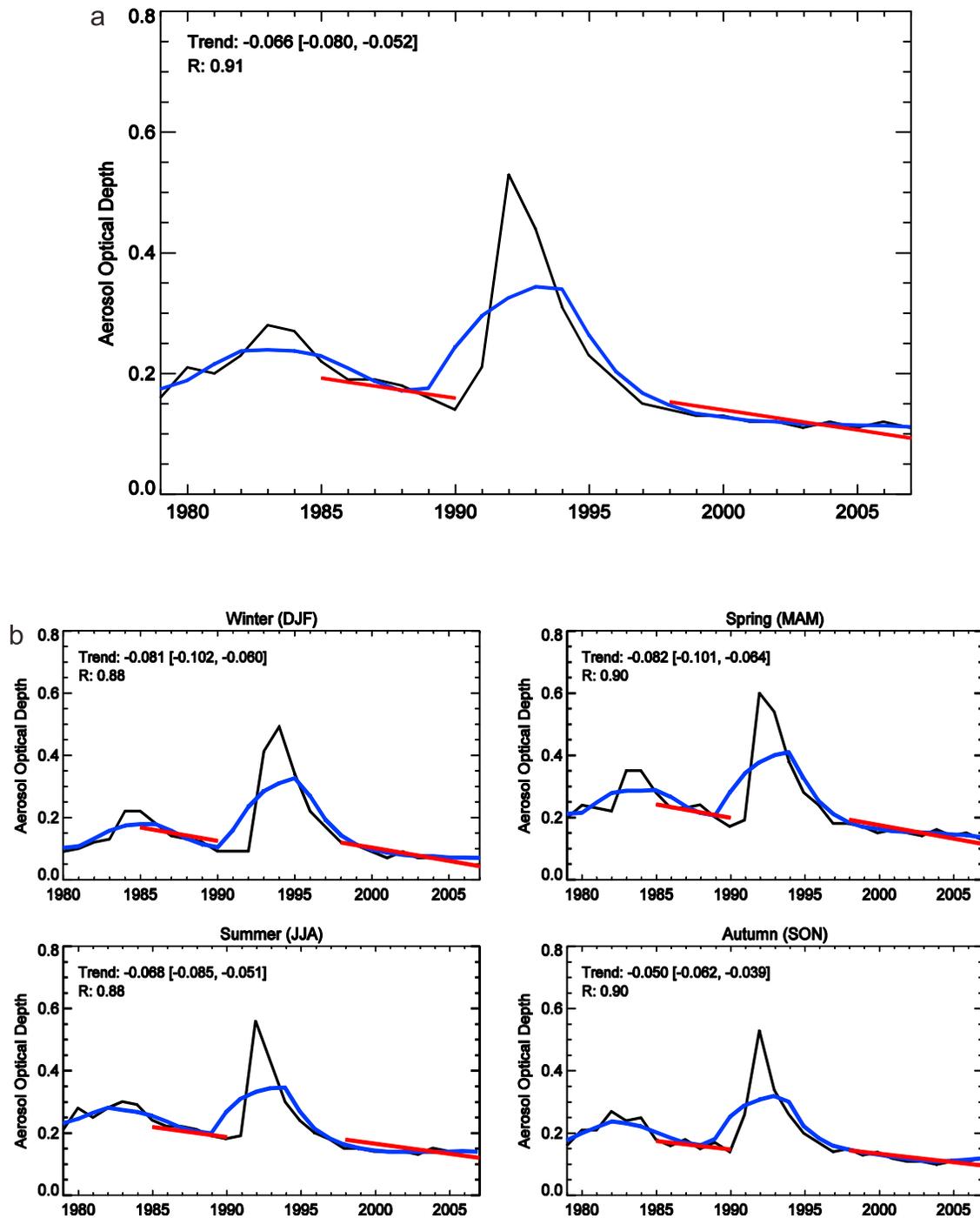


Figure 1. Time series for Europe (a) annual and (b) seasonal mean sulfate aerosol optical depth from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model for the period 1979–2007 (black line). Trends are estimated (per decade) for 1985–2007 from a first-order linear regression model by computing the best fit line (red) with 95% confidence level intervals indicated. A 5 year moving average is applied (blue line). The years 1991–1997 are not included because of the slow decay of the simulated AOD after the Pinatubo volcanic eruption.

in the southern part during summer (Figure 2a). During the period 1998–2007 (Figure 2b), this pattern was also seen during spring. There is also an additional but weaker dipole structure during winter and autumn with positive significant values in the north and negative nonsignificant values in the

south. Moreover, in general the dipole structures were not oriented with a perpendicular north and south line but were instead oriented with a tilt about 45° toward the east.

[18] To further investigate the relationship between the sulfate AOD and NAO, we applied principal component

Table 2. Change in Percent of Annual and Seasonal Mean Sulfate and Total Aerosol Optical Depth (AOD) Over the Period 1985–2007 in Different Regions From the Goddard Chemistry Aerosol Radiation and Transport (GOCART) Model^a

Region	Annual Sulfate/Total	Winter (DJF) Sulfate/Total	Spring (MAM) Sulfate/Total	Summer (JJA) Sulfate/Total	Autumn (SON) Sulfate/Total
Europe	–69/–41	–115/–69	–69/–39	–60/–39	–59/–37
Northern Europe	–73/–46	–132/–77	–76/–46	–57/–42	–64/–44
Southern Europe	–63/–35	–99/–54	–61/–30	–60/–33	–57/–34
Western Europe	–65/–33	–129/–70	–67/–34	–48/–26	–60/–35
Eastern Europe	–75/–50	–111/–70	–76/–49	–71/–53	–64/–42

^aValues denote statistical significance at the 95% confidence level.

analysis. Among all regions, the aerosols explained a significant amount of variability in the NAO for the first component with winter and spring exhibiting the highest values followed by summer and autumn. For Europe, in the detrended time series during 1979–1990, variances of 69% and 77% were seen in winter and spring, respectively. A maximum value of 87% was shown in spring in eastern Europe for the period 1998–2007 in the detrended series. Overall, in both periods (1979–1990 and 1998–2007), this analysis showed a high variance for the first component with a maximum in eastern Europe in winter and spring followed by northern and southern Europe in summer and autumn.

[19] The CWT for the monthly mean sulfate AOD in southern Europe shows the dominant variability in Figures 3a and 3b for the 1979–1990 and 1998–2007 periods. All regions showed a statistically significant power signal around the 1 year period (12 months) for both time spans, signifying a strong stationary annual cycle, but none were as pronounced as the one for southern Europe. Hence, we chose to focus the rest of our investigation of the AOD sulfate and NAO relationship on this region. The CWT during 1979–1990 for the NAO (Figure 3c) exhibited significant power signals at about 4 months during 1984, 6 months during 1986–1987, and a stronger one with the same period from about mid-1988 to 1990. A power signal, though non-significant and less apparent than the one for sulfate AOD, also occurred around the 1 year period (8–14 months) during different intervals. However, it became significant during 2000–2001, 2002–2003, and 2006–2007, although this period is partially outside the cone of influence (Figure 3d). Other significant periods between 2 and 6 months appear in both time periods; however, these are also intermittent.

[20] Wavelet coherence, which shows common power between the two time series for the periods 1979–1990 and 1998–2007, are shown in Figure 4. An antiphase relationship between sulfate AOD and NAO is found with significant power around 12 months from about mid-1981 to mid-1983 and from about mid-1986 until mid-2003 (Figure 4). However, for the 12 month period it was more pronounced in the late 1980s. The arrows contained inside all the significant coherent regions were phase locked, which suggests a cause and effect between the two series. Other regions also showed an antiphase relationship between the sulfate AOD and NAO. In general, the CWT of the NAO had stronger power signals or was redder than the sulfate AOD and regions with low coherence coincided with low power signals from the CWT.

[21] The analysis of the annual cycle of cross correlation between the sulfate AOD and NAO for the period 1979–

1990 (Figure 5, top) revealed an interesting pattern with a negative statistically significant correlation from about mid-autumn to winter (blue) and positive and significant during spring and summer (red). This result was evident throughout all lead and lag times of the two variables (up to 30 months). There was an indication that the sulfate AOD leads the NAO mainly during the spring and summer up until about 12 months and then lags the NAO after that period. During the winter months the antiphase relationship persisted throughout the whole period. For the period 1998–2007 (Figure 5, bottom), there was a clear recurrent statistically significant pattern of negative correlation with the strongest values appearing on a diagonal (dark blue, $r = -0.5$) between March and October. This indicates a strong seasonality of the AOD in the annual cycle, which persisted over the analysis period. The strong seasonality shows a slight lead of the AOD over the NAO, where March AOD is most strongly correlated with January NAO (-0.6) and October AOD is most strongly correlated with August NAO (-0.42). The winter months, however, showed a weak and non-significant correlation close to zero (white) over most of the annual cycle.

4. Discussion and Concluding Remarks

[22] An assessment was made on the performance of long-term simulated total AOD from GOCART compared with surface measurements of total AOD from six sites in Germany and Switzerland given by *Ruckstuhl et al.* [2008]. The results showed that the model was able to capture most of the decline found in the surface measurements, particularly during 1986–2005 for two sites in Germany (Zingst and Lindenberg). The changes in percentage of the modeled sulfate AOD from these same locations during 1998–2005 were greater than the surface measurements by only 1%. These discrepancies, though small, found in the model simulated data could be due to a number of factors including bias errors introduced from the input of meteorological variables and emission data, scale issues due to comparing point measurements to grid box values, and the mismatch in time periods analyzed for trend detection between GOCART and surface measurements. However, despite these circumstances, the model performs well to simulate decadal changes in AOD, which adds confidence to the results found in our study. In addition to the estimate of the total AOD change for Europe, it revealed that the point measurements used of *Ruckstuhl et al.* [2008] were spatially representative for grid scale changes of total and sulfate AOD. This was especially true for the lower altitude stations below 500 m where model and station values agreed most, which suggests that data net-

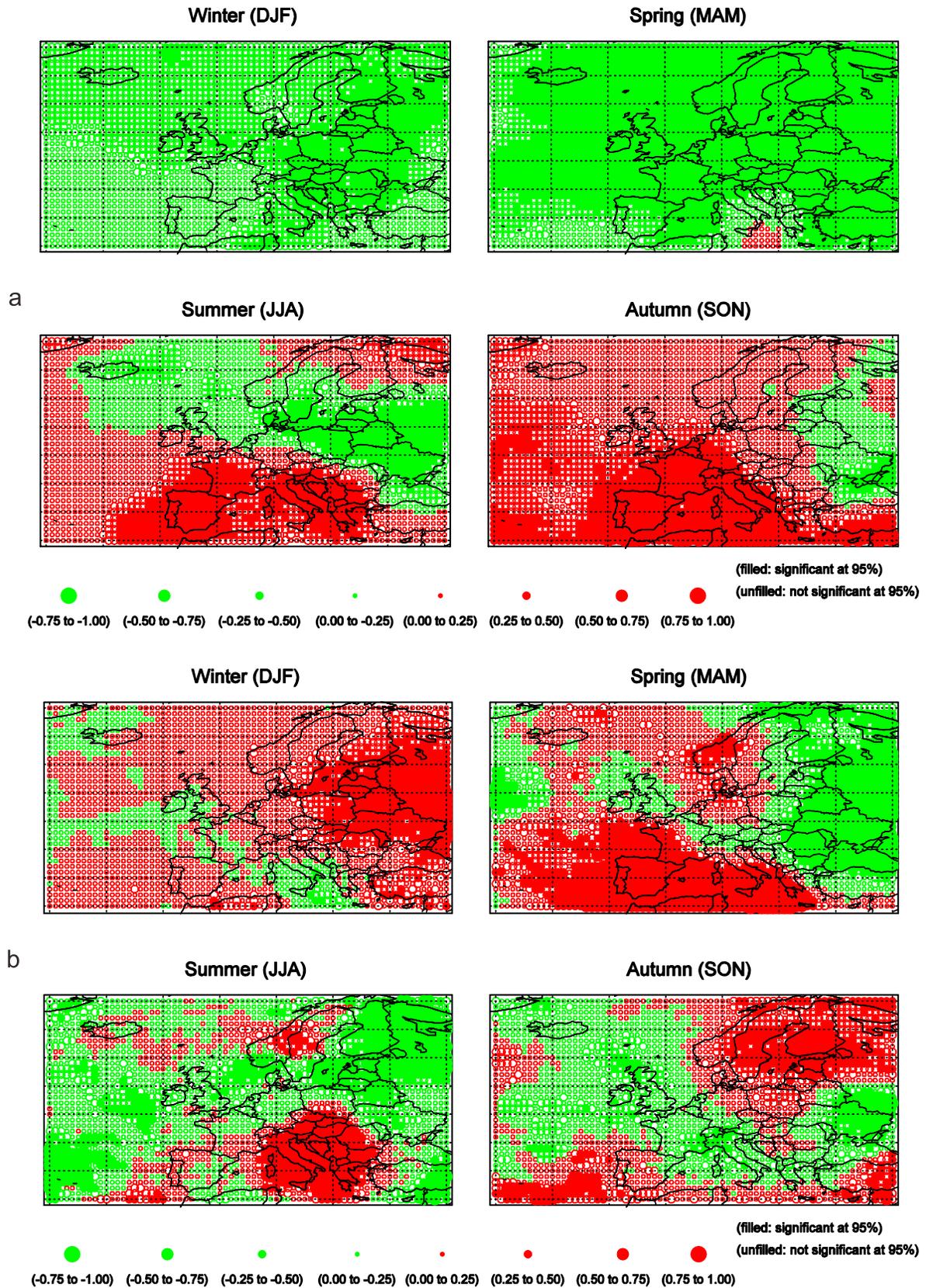


Figure 2. Spatial correlation between seasonal mean sulfate AOD from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model and NAO index for two periods: (a) 1979–1990 and (b) 1998–2007. Significant correlation coefficients at the 95% confidence level are solid circles, and nonsignificant correlation coefficients are open circles.

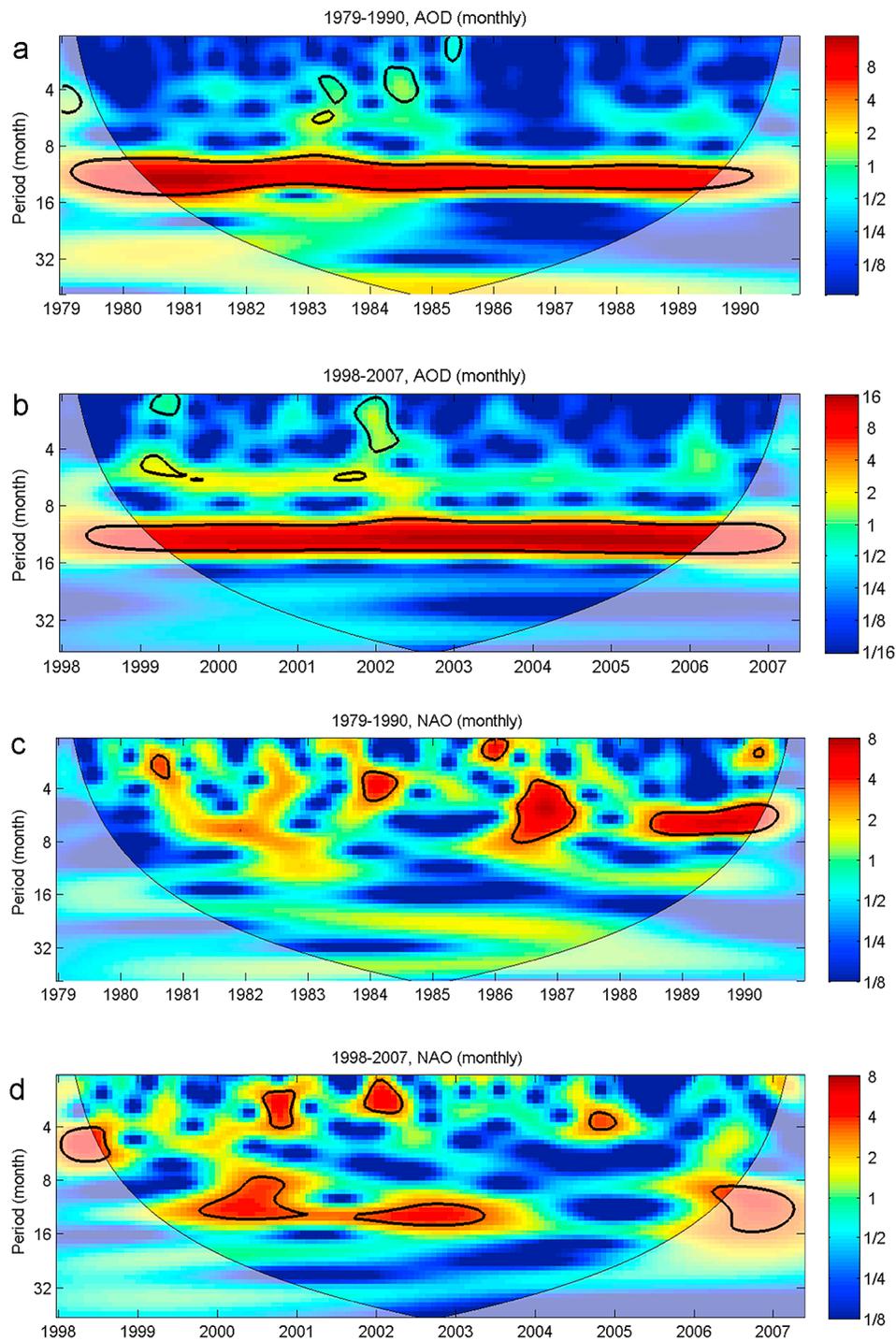


Figure 3. Continuous wavelet transform for southern Europe for (a and b) monthly mean sulfate AOD from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model and (c and d) NAO index for two periods: 1979–1990 (Figures 3a and 3c) and 1998–2007 (Figures 3b and 3d). Thick contour line denotes the 95% confidence level.

works such as these could be useful for representing grid scale variability. At the same time it shows the valuable information gained from aerosol modeling.

[23] The analysis for the annual mean sulfate AOD trends for the period 1985–2007 revealed large significant declines by as much as 69% for Europe with a maximum decline in

eastern Europe of 75%. These changes are similar to the high reduction found in EMEP (European Monitoring and Evaluation Programme) emission inventories of sulfur dioxide (SO_2), a precursor of sulfate through the oxidation process [Chin *et al.*, 2002], in continental and eastern Europe of 65% and 63%, respectively, for the period 1985–2000 [Berglen

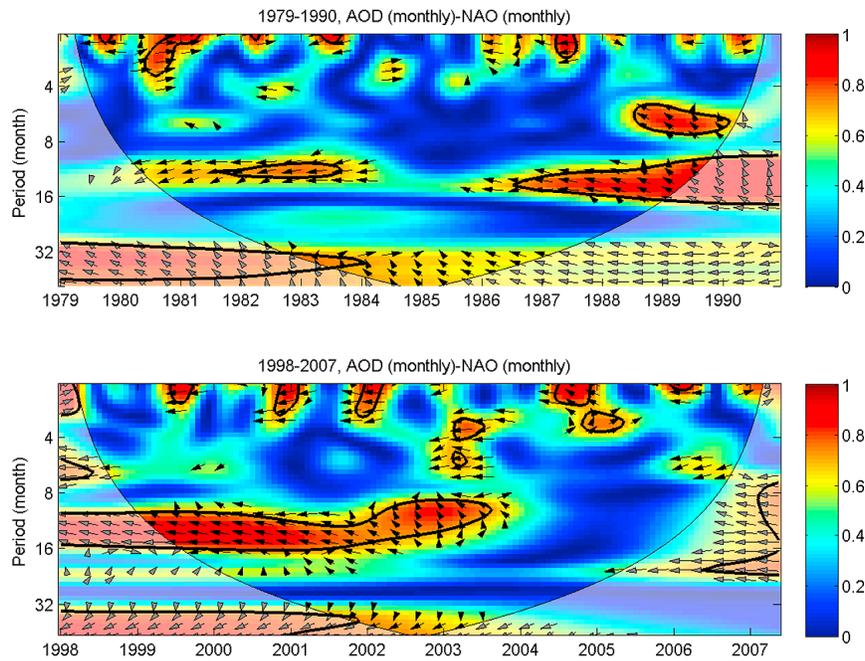


Figure 4. Wavelet coherence for southern Europe between monthly mean sulfate AOD from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model and NAO index for two periods: (top) 1979–1990 and (bottom) 1998–2007. Significance at the 95% confidence level is shown as a thick contour line. Arrows represent the phase relationship between variables (pointing right is in-phase, pointing left is antiphase, pointing up is NAO leading sulfate AOD by 90° , and pointing down is sulfate AOD leading NAO by 90°).

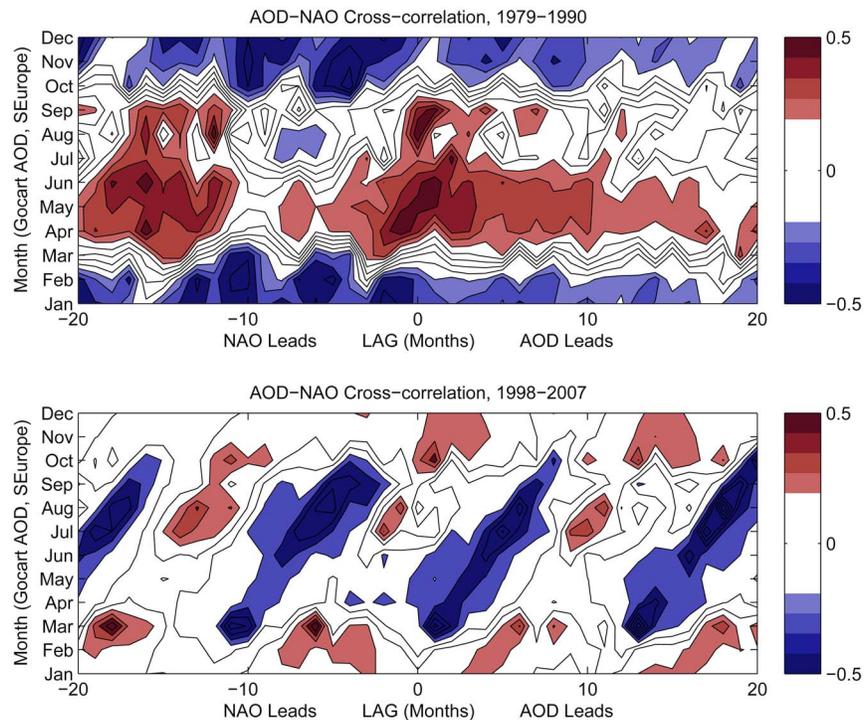


Figure 5. Annual cycle of cross correlation for southern Europe between monthly mean sulfate AOD from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model and NAO index for two periods: (top) 1979–1990 and (bottom) 1998–2007. Statistical significance at the 95% confidence level is indicated in color.

et al., 2007]. Similar results are found by *Vestreng et al.* [2007] also using an emission inventory from EMEP of large reductions of sulfate emission by as much as 54% during 1990–1999 in central and eastern Europe. For the seasonal mean trends in all regions, the greatest decrease occurred in winter and spring followed by summer and autumn. According to the subregions, the maximum decline in AOD was found in northern and eastern Europe followed by western and southern Europe. The larger decline found in these two seasons and subregions are probably due to the greater reduction in burning of brown coal for domestic heating at this time, particularly in eastern Europe [*Marmier et al.*, 2007]. The smaller decline of sulfate AOD found in western Europe from our analysis period of 1985–2007 can be explained by the earlier emission control of sulfate aerosols that was already enforced during the 1980s [*Vestreng et al.*, 2007]. This analysis shows the importance of studying the seasonal variability of the climate in general, which can possibly reveal additional information not observed in the annual temporal scale.

[24] Overall the decrease in long-term sulfate AOD is in line with the emission control of aerosols that came into effect in Europe during the 1980s, where a maximum in emissions occurred in 1988–1989 with a strong decline afterward [*Streets et al.*, 2006]. It also agrees with findings from other studies on the reductions of sulfate compared to our region of study [*Vestreng et al.*, 2007; *Berglen et al.*, 2007] and in the total AOD [*Mishchenko and Geogdzhayev*, 2007; *Ruckstuhl et al.*, 2008; N. Hatzianastassiou *et al.*, manuscript in preparation, 2010] as well as an increase found in the visibility in Europe [*Vautard et al.*, 2009; *Wang et al.*, 2009]. Moreover, this decrease in the long-term sulfate AOD concurs with the rise (brightening) found in the decadal changes of surface solar radiation in Europe after the mid-1980s [*Wild et al.*, 2005; *Norris and Wild*, 2007; *Ruckstuhl et al.*, 2008; *Chiacchio and Wild*, 2010], but only for the annual, spring, and summer mean time scales as found by *Chiacchio and Wild* [2010] and equally by *Sanchez-Lorenzo et al.* [2008] with sunshine duration measurements. This is an important finding, which shows that aerosols on a seasonal basis mainly contribute in spring and summer to the decadal variations in all-sky surface solar radiation in Europe despite maximal reductions in winter. In winter and autumn, however, the long-term decline in sulfate AOD does not agree with the changes reported by *Chiacchio and Wild* [2010] with a near-zero and nonsignificant trend in surface solar radiation in winter for 1970–2000 and a negative significant trend in autumn during the same period. We expect a different result than a decline of sulfate AOD from these results in winter and autumn. However, it strengthens the conclusion from this latter study that the influence of the NAO and its resulting effect on cloudiness could be contributing to the decadal solar radiation changes. At the same time it was further suggested that the strong connection of the solar radiation and the NAO in the low-frequency variability may be due to the interference from aerosols. This places a greater importance on understanding the role of the NAO in all these atmospheric variables. The following discussion develops this argument.

[25] The correlation between the sulfate AOD and NAO for the period 1979–1990 in winter and spring showed good to moderate negative values that were significant, with low

positive nonsignificant values in summer and fair positive significant values in autumn. These results physically imply that during winter, for example, the effect of aerosols on the climate was at a minimum when the NAO was in its strong positive phase in the 1980s. In summer the sulfate AOD and the NAO were in-phase, meaning that during the large positive index of the NAO aerosols played a larger role in affecting the amount of solar radiation reaching the surface. This argument agrees with Figure 3 from *Chiacchio et al.* [2010], where dimming and brightening is seen in summer and a slight positive trend in solar radiation was observed throughout winter. The autumn and spring correlations between sulfate AOD and NAO are not so clear and do not agree with *Chiacchio et al.* [2010]; however, by analyzing this relationship on a monthly time scale, as in the cross-correlation analysis, more information was revealed. To show their correlation spatially during 1979–1990 we have constructed a map that revealed mostly homogeneous negative values across Europe in winter and spring. For summer a dipole pattern emerged tilted at about a 45° angle toward the east with significant negative correlation in the central to northern parts of Europe and significant positive correlation in the south. The spatial map of the correlation for the period 1998–2007, on the other hand, exhibited dipole structures in all seasons with spring most resembling the summer pattern discussed before. These maps during this latter period have a more complex pattern, which is unclear at this point to explain the interaction between aerosols and NAO. Though this type of statistical analysis showed in general a good correlation between sulfate AOD and NAO, additional statistical tests were needed to determine whether a physical link exists between the two variables.

[26] In order to accomplish this, we applied wavelet analysis separately for the sulfate AOD and NAO to show variations of their spectral power as a function of both time and period. We observed 1 year significant band signals seen in the sulfate AOD and NAO that persisted throughout the periods 1979–1990 and 1998–2007. However, the signal was slightly weaker for the NAO but showed an intermittent feature not present in the sulfate AOD. The coherence between the two variables was high and significant in the 1 year period from about mid-1986 to 1990 and from 1998 to mid-2003. Moreover, it was most pronounced in the late 1980s, which signifies the time when sulfate emissions and the NAO index were at their peak. In addition, most regions of significant coherence were found to be phase locked, which according to *Grindsted et al.* [2004] would be needed in order to suggest a cause and effect relationship. This is an important observation because it makes it more likely, instead of just speculating, that their relationship is not merely by chance or that a physical link may exist between the two.

[27] We demonstrated that there exists a link between the southern Europe sulfate aerosols and the NAO showing that they are anticorrelated, which agrees with the study by *Fischer-Bruns et al.* [2009], where an anticorrelation is found between the anthropogenic sulfate aerosols and the NAO in future climate simulations for the North Atlantic. We further revealed an antiphase relationship that was significant from about midautumn to winter and an in-phase significant relationship in spring and summer. This result showed additional information not seen in the wavelet analysis where only a strong antiphase relationship was observed,

signifying the winter dominance of the NAO. Instead, the year-long presence of this mode of variability including the summer [Hurrell *et al.*, 2003; Folland *et al.*, 2009; Sun *et al.*, 2009] was detected in the cross-correlation test with opposite effects apparent between seasons. Moreover, because the NAO during 1979–1990 was primarily in its positive phase [Hurrell *et al.*, 2003], we propose that sulfate aerosols in winter and autumn were acting at a minimum on the surface solar radiation according to our finding that sulfate aerosols and NAO are anticorrelated with one another. This agrees with the suggestion of the larger role that the NAO and associated cloudiness have on the decadal solar radiation changes in winter and autumn after the mid-1980s. With a positive correlation between sulfate aerosols and NAO in the spring and summer months, aerosols had a larger influence on the long-term solar radiation variations, which is reflected in Figure 3 of Chiacchio and Wild [2010], where a dimming and brightening from the all-sky surface solar radiation is observed in these two seasons. In addition, from the cross-correlation analysis, the annual cycle of the lags during 1979–1990 showed that from about 0 to 12 months sulfate aerosols were leading and then lagging by the NAO afterward. From this result, it is unclear if the sulfate aerosols are forcing the NAO. This was also the case for the period 1998–2007 without a clear indication of which variable is leading. However, it can be inferred that because the NAO was at a weaker positive phase in this period, the interactions with other atmospheric variables were not as strong.

[28] We have shown that a strong relationship exists between sulfate aerosols and the NAO particularly in the 1980s. However, the results from the statistical techniques used in this study do not resolve the argument of whether aerosols have any influence on the atmospheric circulation and vice versa. It is also interesting to note that this period coincides at the same time when there was an extraordinary increase of emissions of anthropogenic sulfate aerosols with a peak in 1988–1989 [Streets *et al.*, 2006], possibly suggesting a forcing on the NAO by these aerosols. Luo *et al.* [2009] presented a conceptual model on the thermodynamic and dynamic impact of tropospheric aerosols on SSTs, mainly over the subtropical Atlantic Ocean, and its resulting effect on the phase change of the NAO. In light of this study, we propose that it might be possible that the decadal changes reported here of sulfate aerosols could also act in the same manner and partially contribute to the large positive trend in the NAO during the 1980s. Moreover, our study also complements the findings of those that investigate aerosol forcing and its impact on atmospheric dynamics including a reduction in the baroclinicity of the atmosphere, changes in stationary wave patterns, and expansion of the Hadley cell [e.g., Kim *et al.*, 2007; Zanis, 2009; Allen and Sherwood, 2010; Lewinschal and Ekman, 2010] with the implication that regional aerosol radiative forcing can cause changes in large-scale circulation such as the NAO. In our paper we have provided new information on the phase relationship between tropospheric anthropogenic sulfate aerosols and the NAO with a seasonal dependence and we have helped explain dimming and brightening on a seasonal time scale in Europe. However, it is still inconclusive whether large changes in anthropogenic sulfate aerosols influence low frequency NAO variations, in particular the large positive NAO trend

observed in the 1980s [Visbeck *et al.*, 2001; Bojariu and Gimeno, 2003]. More research is needed to clearly demonstrate this and the underlying physical mechanisms involved, which may be feasible with additional statistical techniques or modeling efforts. Such evidence could ultimately lead to a better understanding of the interaction of anthropogenic aerosols and NAO and their role in extreme climate events in Europe including droughts and heat waves and possibly enhance better prediction of the NAO.

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- M. Chiacchio, T. Ewen, and M. Wild, Institute for Atmospheric and Climate Science, ETH Zurich, Universitaetsstrasse 16, CH-8092 Zurich, Switzerland. (marc.chiacchio@env.ethz.ch)
- M. Chin and T. Diehl, NASA Goddard Space Flight Center, Greenbelt, MD 20771, USA.