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Have aerosols affected trends in visibility and precipitation in Europe?

Camilla W. Stjern,[1] Andreas Stohl,[2] and Jón Egill Kristjánsson[1]

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[1] Here we investigate whether the large variations in anthropogenic aerosol concentrations over the last decades have had a notable effect on precipitation in Europe. Our main focus is on the heavily industrialized region formerly known as the Black Triangle (BT), where pollution levels increased until the late 1980s and then decreased substantially. Precipitation changes in this area are compared to changes in a clean coastal region in western Europe with minor pollution trends but with potentially higher aerosol-precipitation susceptibility. We find that pollution reductions caused a substantial increase in horizontal visibility of 15 km from 1983 to 2008 in the BT, contrasting a 2.5 km increase in the clean region. Over the same period total precipitation trends show no sign of aerosol influence, neither in years of emission increases nor emission reductions, and a circulation-based precipitation estimate indicates that the observed trends may be entirely explained by atmospheric circulation changes. The annual frequency of light precipitation events, however, increased significantly in the clean region, and in the BT we find significant changes in both total and light precipitation frequency for wind directions at which sulphate trends were largest. The trends were most pronounced in the summer season. Altogether, we find no verification that changes in pollution have caused measurable changes in total precipitation in Europe, but changes in light precipitation types bear a potential aerosol signal.

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1. Introduction

[2] The ability of sulphate particles (created by gas-to-particle conversion from sulphur dioxide) to influence clouds was known as early as the 19th century, when Scottish scientist John Aitken recommended restrictions on sulphur emissions to reduce the frequency of the disturbing London fogs [*Aitken*, 1880]. Since then, research has brought compelling evidence that an increase in aerosol concentrations is conducive to increased cloud droplet numbers and decreased cloud droplet radii [*Twomey*, 1977]. While this clearly influences the radiative properties of clouds, the magnitude and even the sign of the aerosol effect on precipitation formation is less clear [*Stephens and Feingold*, 2009; *Khain*, 2009; *Levin and Cotton*, 2008].

[3] *Khain* [2009] argued that the net effect of aerosols on precipitation depends on the relationship between condensate generation (droplet condensation and ice deposition) and condensate loss (evaporation and sublimation). He found that the main factors affecting this relationship are cloud type, air humidity, and wind shear. The fact that the aerosol effect on precipitation depends on meteorological conditions is reflected in the large spread in results from observations and model studies: In warm clouds, reduced cloud droplet sizes tend to lower the collision-coalescence efficiency between the droplets, thus slowing down precipitation-generating mechanisms [*Albrecht*, 1989; *Gunn and Phillips*, 1957]. Some studies [e.g., *Rosenfeld*, 2000; *Qian et al.*, 2009], although debated [*Alpert et al.*, 2008; *Ayers*, 2005; *Ayers and Levin*, 2009], indicate that in shallow clouds, this effect has a potential to shut off precipitation altogether. For instance, *Givati and Rosenfeld* [2004] found that orographic precipitation suppression near industrialized areas can amount to 15–25% of the annual precipitation sums. Less clear is the effect of delayed precipitation onset in deep convective clouds, where transport of cloud water by updrafts to supercooled regions may release additional latent heat of freezing and result in more vigorous precipitation events [*Andreae et al.*, 2004]. However, precipitation invigoration in convective clouds mainly seems to occur when cloud bases are sufficiently warm [*Rosenfeld et al.*, 2008; *Bell et al.*, 2008], and studies of cooler convective clouds show suppression also here [e.g., *Teller and Levin*, 2006; *Khain et al.*, 2001]. *Khain* [2009] noted that the aerosol-precipitation effect in deep convective clouds is highly dependent on the relative humidity of the surroundings, arguing that precipitation enhancement will only occur in moist maritime environments, where evaporation is relatively inefficient. Similarly, *Fan et al.* [2009] found through model studies a strong dependence on wind shear, and observed convective precipitation enhancement by aerosols only in cases with weak vertical wind shear, as strong

[1] Department of Geosciences, University of Oslo, Oslo, Norway.
[2] Norwegian Institute for Air Research, Kjeller, Norway.

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wind shears were associated with more detrainment and thus more evaporation.

[4] Other aerosol types such as giant cloud condensation nuclei (CCN), of which for instance sea salt is a major source, have been shown to enhance precipitation [*Johnson*, 1982]. In a model study by *Teller and Levin* [2006], however, this enhancement only partially offset the precipitation suppression of cool convective clouds by regular-sized CCN. The same work found reduced precipitation from cool-base convective clouds under addition of more ice nuclei, while in shallow supercooled clouds this would tend to enhance precipitation due to glaciation and the Bergeron-Findeisen effect [e.g., *Wallace and Hobbs*, 1977]. Cloud chamber studies [*Gorbunov et al.*, 2001] have shown a potential, particularly at low temperatures, for ice crystals to form on soot particles, which are emitted in abundance particularly from low-technology coal-fired power plants [*Wehner et al.*, 1999]. This tendency of aerosols to affect precipitation processes differently in different types of clouds and environments illustrates the nonlinearity and complexity of aerosol-cloud-precipitation interactions; after more than a hundred years of research in cloud physics, the indirect effects of aerosols on precipitation are still poorly understood.

[5] In addition to the indirect aerosol effects, urban areas may also influence clouds by other pathways. Decreasing aerosol concentrations cause increased surface absorption as well as a vertical redistribution of heat, which may affect the atmospheric stability and hence cloud formation [*Rosenfeld et al.*, 2008]. Moreover, changes in surface properties by growing cities or changes in land use may affect convection and precipitation through a variety of mechanisms, as summarized by *Shepherd* [2005]. For instance, *van den Heever and Cotton* [2007] found that urban-forced convection downwind of an urban area, rather than the presence of aerosols, influenced whether convective systems developed or not. Finally, it should be noted that while aerosols affect precipitation through direct and indirect effects, precipitation will also affect aerosol concentrations by wet deposition removal.

[6] In the present paper, we examine whether a large change in the emission of anthropogenic aerosols and their precursors can modify cloud microphysical properties in such a way as to cause measurable variations in precipitation. Europe provides a natural background for this analysis, with periods of large emission changes coinciding with the availability of long precipitation time series in a relatively dense station network. The detection of aerosol signals in precipitation time series has been much debated, and while some find a strong aerosol-precipitation link [e.g., *Givati and Rosenfeld*, 2004; *Jirak and Cotton*, 2006; *Rosenfeld*, 2000], others find weak or even no effects [e.g., *Alpert et al.*, 2008; *Ayers*, 2005; *Ayers and Levin*, 2009; *Halfon et al.*, 2009; *Schultz et al.*, 2007]. Searching for a potentially small aerosol signal in precipitation measurements presents a challenge, as natural variability in precipitation is large, and as aerosol effects may vary even in sign depending on the conditions, as accounted for above. To increase our chances of detecting a potential signal, we therefore focus on a region in central Europe formerly known as the Black Triangle (BT), where variations in aerosol concentrations have been particularly large and abrupt. Here, pollution levels increased up until the late 1980s and decreased dramatically thereafter. For this area, *Krüger and Graßl* [2002] found a pronounced decrease in cloud albedo in periods of decreasing aerosol concentrations, indicating an aerosol effect on cloud droplet sizes according to the theory of *Twomey* [1977]. As a comparison, motivated by earlier findings [e.g., *Andreae et al.*, 2004; *van den Heever and Cotton*, 2007] that regions with lower background concentrations may be more susceptible to aerosol-precipitation interactions, we also study precipitation changes in a clean region at the west coast of Europe, where trends in pollution have been minor. To account for opposing aerosol effects depending on meteorological conditions, we stratify the data according to precipitation type, season, cloud type, wind direction, and weather type.

2. Data and Methods

[7] A summary of all the data sets used in the present paper is given in Table 1. To study precipitation in Europe we used synoptic weather observations (abbreviated SYNOP) for 0000, 0600, 1200 and 1800 UTC from the European Centre for Medium-Range Weather Forecasts' (ECMWF) Meteorological Archive and Retrieval System, for which data were available for the period 1983–2008. The SYNOP stations are marked as red dots in Figure 1. Only data from stations with complete time series for the 1983–2008 period were included, which gave us a total of 30 stations from the BT area and 18 stations from a less polluted area on the west coast of Europe, which will henceforth be referred to as "the clean region". A study by *Goswami et al.* [2006] showed the need for studying large areas to be able to discern precipitation trends reliably, as opposed to studying single stations which will be more prone to large variability and sampling issues. The trends in the present analysis are therefore calculated from areal mean precipitation based on the above mentioned stations.

[8] While the SYNOP station data have the advantage of daily temporal resolution and a large number of meteorological variables, gridded data allow for analyses of spatial variations in trends. We therefore use gridded precipitation data sets from the Climate Research Unit (CRU) of the University of East Anglia and from the Global Precipitation Climatology Project (GPCP) as a supplement to our analyses. Additional advantages are that these data go further back in time, and have undergone corrections and homogenizations that the raw SYNOP data from our source have not.

[9] The CRU TS 3.0 data set contains monthly precipitation sums between 1901 and 2006 on a 0.5° global grid, and is based on interpolated surface observations which are homogenized [*New et al.*, 2000]. For the CRU data set, the BT region comprises 50 grid cells. Meanwhile, the GPCP Version 2.1 Combined Precipitation Data Set contains monthly means of daily precipitation sums based on a combination of satellite and gauge measurements in a 2.5° global grid [*Adler et al.*, 2003]. The gauge measurements are homogenized as well as corrected for systematic errors (using bulk correction factors for monthly climatological conditions as accounted for in *Adler et al.* [2003]) such as precipitation under catch due to aerodynamic effects, which is a particularly large problem at high-altitude sites. The BT area comprises only two GPCP grid cells due to the larger grid sizes. The area extracted from the gridded data sets is marked as a red square in Figure 1.

Table 1. Overview of the Data Sets Used

Data Set	Period	Source	Spatial Resolution	Temporal Resolution	Stations or Grid Cells	Area
SYNOP, BT Area	1983–2008	Surface stations	–	Daily	30	49.50°N to 52.00°N 12.00°E to 18.00°E
SYNOP, Clean Region	1983–2008	Surface stations	–	Daily	18	44.50°N to 49.50°N −5.00°E to −0.50°E
CRU, BT Area	1901–2006	Surface stations	0.5° × 0.5°	Monthly	50	50.00°N to 52.50°N 12.50°E to 17.50°E
GPCP, BT Area	1983–2008	Surface stations and satellites	2.5° × 2.5°	Monthly	2	50.00°N to 52.50°N 12.50°E to 17.50°E
EMEP emissions	1980, 1985, 1990–2007	Country emissions as used in EMEP models	–	Annual	–	–
EMEP measurements	1983–2008	Surface stations	–	Daily and monthly	12	–
GSOD	1973–2008	Surface stations	–	Monthly	56	–

[10] Measurement data of sulphate in aerosols from the European Monitoring and Evaluation Program (EMEP) were used to get an indication of the spatial and temporal variations of sulphate concentrations in Europe. We used both daily data values, sorted into country averages to present regional differences in the trends in Europe, as well as monthly means from a selection of 12 stations with particularly long and consistent time series and high data credibility (W. Aas, personal communication, 2008). Most of the 12 stations were located in central Europe; see green dots in Figure 1.

[11] Finally, a comparison of horizontal visibility changes in different regions of Europe was performed using synoptic station data from the Global Surface Summary of the Day (abbreviated GSOD), which includes daily mean surface observations from the Global Station Network. Data were downloaded from the NOAA National Data Center Climate Data Online (http://www7.ncdc.noaa.gov/CDO/cdo). We selected stations from the BT and clean region, as well as from two other relatively polluted and two relatively clean regions, respectively.

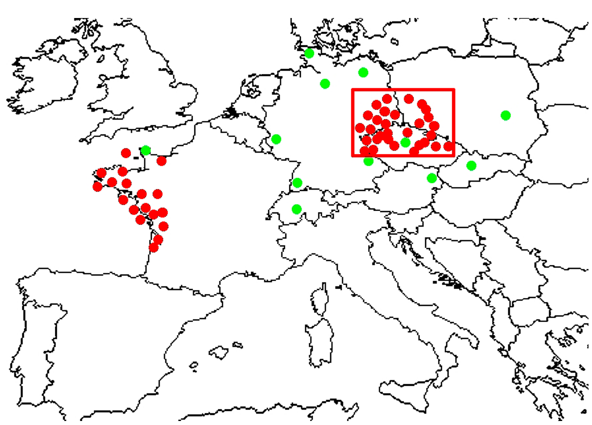

Figure 1. Red dots mark the position of the meteorological stations from which precipitation data are used in the analysis. The Black Triangle (BT) region is the rightmost region on the map. The area on the west coast of France is referred to as the "clean region". Green dots mark the position of the EMEP stations from which sulphate data are used, and the red square marks the BT area for which gridded precipitation data were extracted.

[12] To extract trends from the data, we use the non-parametric Mann-Kendall test, in which no assumption for normality is required [*Yue et al.*, 2002]. When indicating that trends or correlations are statistically significant at the 5% or 1% level, we refer to p values lower than 0.05 or 0.01, respectively. When no information of significance level is given, the trend or correlation in question is not statistically significant at any of the above levels. Unless otherwise stated, percentage trends are calculated relative to the mean of the period in question.

3. Study Area

[13] The heavily industrialized Black Triangle is an area named from the triangle of the meeting borders of Germany, Poland, and the Czech Republic (see the rightmost cluster of stations in Figure 1). This area, home to Europe's largest basin of lignite coal, hosted numerous coal-fired power plants with high levels of sulphur emissions [*Blas et al.*, 2008]. Suffering excessive concentrations of sulphur dioxide, the BT experienced severe forest dieback in the 1970s and 1980s: based on satellite observations, *Ardo et al.* [1997] found that 50% of the coniferous forest in this area disappeared between 1972 and 1989. Following the political and economical changes in 1989, emissions declined markedly, fulfilling our need for large and abrupt aerosol concentration trends. While changes in aerosol concentrations do not necessarily imply changes in CCN concentrations of similar magnitudes, direct measurements of CCN concentrations were not available to us. However, based on the knowledge that sulphate aerosols generally are efficient CCN [*Aitken*, 1880] and that strong correlations are observed between aerosol optical thickness and CCN concentrations [*Andreae*, 2009], we assume that the dramatic emission reductions have at least lead to a substantial reduction in CCN concentration.

[14] Topographic barriers in the BT area are the Sudeten mountain range in the east (on the border between Poland and the Czech Republic) and the Ore Mountains in the west (on the border between Germany and the Czech Republic). The annual mean precipitation in the area is 530 mm, and monthly precipitation sums peak during summer and are lower during winter. The annual precipitation sum varies between the 30 BT stations, with a standard deviation of 97 mm. For summer (defined as June through August) the regional monthly mean is 62 mm with a 17 mm standard

deviation between the stations, whereas for winter (defined as December through February) the monthly mean is 37 mm and the standard deviation is 14 mm. While summers are dominated by convective clouds and convective precipitation, here identified by observed rain showers and heavy precipitation events, winters have more observations of light precipitation and drizzle as well as stratiform clouds. We define light precipitation as events with less than 0.5 mm in 12 hours and heavy precipitation by more than 10 mm in 12 hours. The highest precipitation amounts occur in summer, but the number of days with precipitation is approximately the same in summer and winter, which can be attributed to the fact that the summer precipitation events typically are more intense. The governing wind direction over the area is westerly.

[15] We also study a relatively clean region on the west coast of Europe, more specifically France (see leftmost cluster of stations in Figure 1). Here, lower levels of industrial activity as well as a governing westerly wind carrying maritime air to the region have caused the initial pollution loads and consequently pollution trends to be small. From a field experiment in central Germany, *Dusek et al.* [2006] reported mean 6-hourly CCN concentrations of around 3000 cm^{-3} for air coming from the Ruhr region, which like the BT is characterized by numerous coal-fired power plants, and concentrations around only 1000 cm^{-3} for air coming from the west and hence over the clean region.

[16] Climatological differences preclude direct comparison between the two regions. Whereas monthly precipitation sums are highest during summer in the BT, the highest precipitation amounts in the clean region come in the wintertime. Also, the frequency of light precipitation events is fairly equal in the BT and the clean region, but the clean region is marked by twice the annual number of heavy precipitation events as the BT. Similarly, there may be important dissimilarities in the characteristics of the aerosols in the two regions; for instance, the clean region is likely to contain a higher number of giant CCN such as sea salt, which do not affect cloud microphysics in the same way as sulfate. Even so, due to different precipitation susceptibilities, it is of interest to see how precipitation changes have differed between the two regions.

4. Sulphate Trends

[17] At the commencement of the 20th century, European industrialization initiated increasing atmospheric concentrations of sulphur dioxide. Abatement measures motivated by degraded urban air quality as early as in the late 1800s were at the time met with skepticism as the sulphuric fogs were believed to have disinfecting capacities, supposedly lowering death rates [*Thorsheim*, 2006]. Emissions kept increasing and culminated in the 1970s, when dying forests and damaged water systems due to acid rain sparked a continental effort to reduce pollution. This contributed strongly to reduce European sulphur dioxide emissions, which were cut by 73% between 1980 and 2004 [*Vestreng et al.*, 2007].

[18] While mean emission levels in Europe were decreasing in this period, there were distinct geographical variations in the timing. Reductions in western Europe started in the 1980s, whereas emissions in eastern Europe (defined in the present article as the area around the BT) were strongly reduced only after the political and economic breakdown in the region in the late 1980s [*Vestreng et al.*, 2007]. Figure 2a shows SO_x emissions for eastern European countries corresponding to the BT (Poland and the Czech Republic) and for western European countries corresponding to the clean region (Ireland, France, and Spain), based on EMEP emission data up to 2007. Figure 2b shows measurements of sulphate in aerosols for the same countries. Figure 2 confirms that concentrations in the eastern countries have a later and more dramatic decrease than in the western countries; compared to the beginning of the time series, emission levels (concentrations) decreased by 72% (59%) for western countries and by 90% (82%) for eastern countries for the period shown.

[19] Other chemicals may also act as CCN precursors, for instance nitrates (at least when compared to circumstances where no condensable vapors other than water were present [*Kulmala et al.*, 1993]) and ammonia [e.g., *Ishizaka and Adhikari*, 2003]. The European emissions of nitrates and ammonia did also increase over the course of the 20th century [*van Aardenne et al.*, 2001], but emission reductions over the past three decades were not as dramatic for these pollutants as for sulphur, and even less so in western than in eastern Europe. Summing the emissions of SO_x, NO_x, and NH_3 for the same countries as in Figure 2, we find that in eastern Europe the largest contribution to the "total" emission comes from SO_x, while for western Europe the largest contributor is NO_x. For the total emission of these CCN precursor gases, we therefore find an even larger difference between east and west; compared to the beginning of the time series, emission levels decreased by 44% for western countries and by 82% for eastern countries for the period shown.

5. Visibility Trends

[20] The overall emission reductions since the 1980s and the consequential recovering atmospheric transparency can be recognized in measurements of clear-sky solar radiation in Europe. After decreasing substantially since the 1950s, these measurements show strong increases after the 1980s; a phenomenon commonly referred to as global brightening [*Alpert et al.*, 2005; *Wild*, 2009]. Such a recovery should be noticeable in measurements of horizontal visibility. Therefore, lacking a dense aerosol measurement network in our areas of interest, we investigate whether there have been any changes in mean visibility that could demonstrate the nature of the aerosol variations in these specific regions.

[21] Figure 3 shows horizontal visibility measurements from selected stations in eastern and western Europe, based on GSOD data. In concordance with the geographic difference in the sulphate trend shown in Figure 2, eastern areas have had a much larger visibility improvement than western areas, and here the change also comes later. Supporting these findings, *Vautard et al.* [2009] showed that the frequency of low-visibility events in Europe has decreased by more than 50% over the past 30 years, and that there is a significant correlation between SO_2 emission reductions and the decrease in the number of low-visibility events.

[22] To illustrate this close connection between sulphate concentrations and horizontal visibility for the BT area specifically, we compare the mean sulphate concentration from a selection of EMEP stations (mainly in central Europe, green dots in Figure 1) to the mean horizontal visibility for

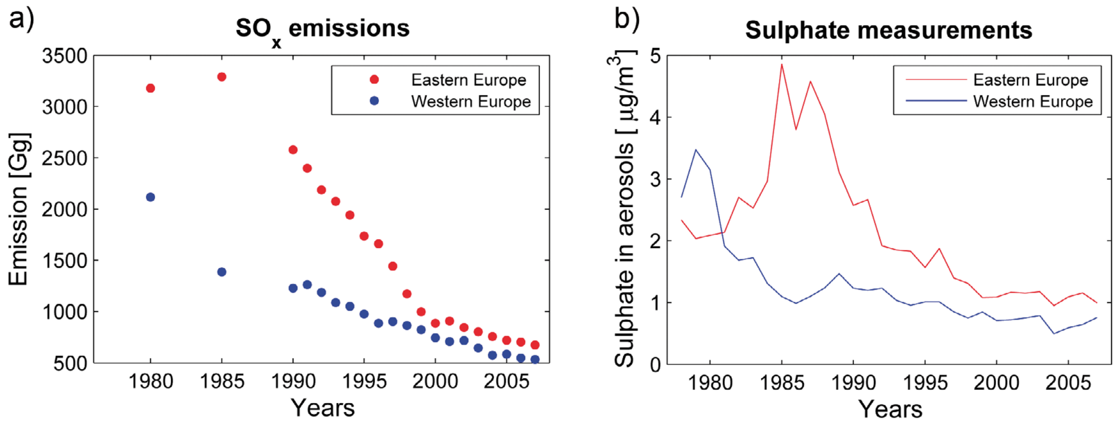

Figure 2. (a) Country mean SO_x emissions for the years 1980, 1985, and 1990–2007, and (b) mean measurements of sulphate in aerosols for 1978–2007 from selected countries in western Europe (France, Spain, and Ireland) and eastern Europe (Poland and the Czech Republic), based on EMEP emission and measurement data, respectively.

the 30 SYNOP stations in the BT for which we have precipitation data. To avoid variations in fog frequency affecting the visibility trend we excluded days with observations of fog from the visibility analyses. Figure 4 shows that while mean concentrations of sulphate from our chosen EMEP stations were two and a half times higher in 1983 than in 2008 (corresponding to a 72% decrease from 1983 values), the mean horizontal visibility in the BT for the same period increased by about 15 km or 134% compared to 1983 values. Both trends were significant at the 1% level, and the Pearson's correlation coefficient between the two variables is $r = -0.94$ (also significant at the 1% level). Figure 4 demonstrates not only opposite trends, but also coherence in interannual variations: The cold winter of 1996 with stagnant air probably caused peak sulphate concentrations, and this is recognized as degraded visibility in Figure 4. A somewhat earlier decrease in sulphate than in visibility can be explained by the fact that the EMEP stations (see green dots in Figure 1) are located mostly outside the BT area where mitigation efforts were taken earlier (as shown in Figure 2). Ultimately, we regard the large visibility trend in the BT area as an indication that the aerosol trend in this region was at least as strong as in the larger area from which the sulphate data shown in Figure 4 were obtained.

6. Variations in Precipitation

6.1. Total Precipitation

[23] As a first approach we want to see whether there have been any distinct trends in precipitation amounts in the periods of large sulphate variations. We need to study typically warm and typically cold conditions separately since the aerosol effect on precipitation may vary in both magnitude and sign depending on the conditions, as explained in section 1. In addition to annual precipitation sums, we therefore analyze winter and summer precipitation separately.

[24] Between 1983 and 2008 annual precipitation in the BT had a statistically insignificant (p value 0.2) increase of 13% with respect to mean annual precipitation (see Figure 5 and Table 2). The exact trend varied between the individual stations; 20 of the 30 stations experienced a precipitation increase of more than 10%, but at only 3 of the stations was the increase statistically significant. The clean region had a decrease of 17% (p value 0.2) in the annual precipitation

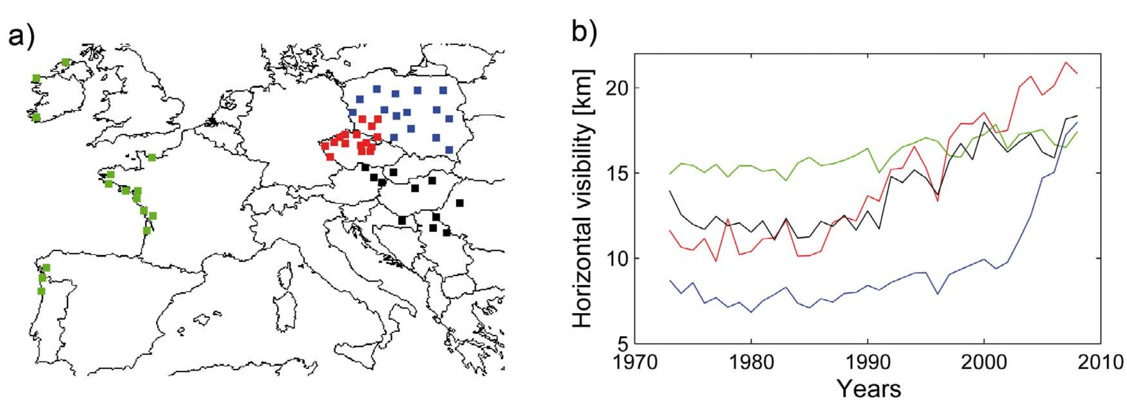

Figure 3. Horizontal visibility from 1973 to 2008 for a selection of stations from GSOD. The stations are divided into different regions, and the annual mean visibility of the stations represented by a specific color of dots in Figure 3a is displayed in Figure 3b.

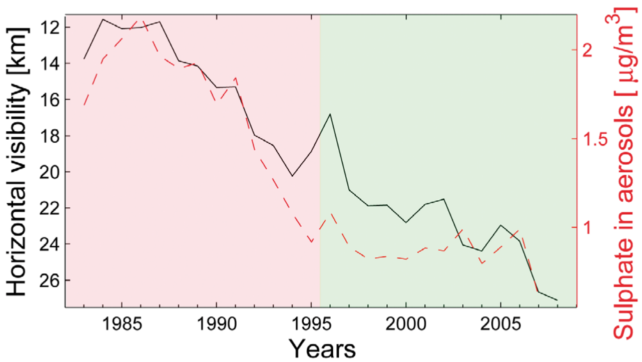

Figure 4. Annual mean values of sulphate in aerosols for central Europe (see locations of the EMEP stations in Figure 1) and mean horizontal visibility (excluding days with fog) for SYNOP data from the BT area; note the reversed visibility axis. The period with the largest decrease in sulphate concentrations (1983–1995) and the subsequent period with a small sulphate trend (1996–2008) are marked with different background colors.

sum. As can be seen in Table 2, precipitation increased in both seasons in the BT, but decreased in winter and increased in summer in the clean region. Interestingly, the annual number of days with precipitation (see "Precipitation days" in Table 2) showed a significant increase in the clean region, in spite of a decrease in precipitation amounts. The BT saw an insignificant increase also in this parameter.

[25] As Figure 4 showed, the 1983–2008 period can be divided into a period of strong reduction in sulphate concentration (1983–1995) and a subsequent period of comparatively little change (1996–2008). In the BT, the largest precipitation increase (+17%) occurred in the period of pollution reduction, followed by a mere 2% decrease in the period of small pollution changes. The clean region, on the other hand, saw a 4% decrease in the first period and a 10% decrease in the second period. It should be noted that while there does not seem to be a systematic tendency of trends being largest in the pollution reduction period, these trends are highly statistically insignificant.

[26] To ensure that the change in the SYNOP data is representative of the period and region in question, we compare the SYNOP precipitation trend for the BT area to trends based on data from CRU and GPCP, using the 1983–2006 period for which data are available from all three sources. For the clean region, the distribution of the 18 stations did not match the large GPCP grid cells well, so only a comparison between SYNOP and CRU data was performed and showed very similar precipitation amounts (not shown). A comparison between the three datasets for the BT region can be seen in Figure 6. Discrepancies are not unexpected as the SYNOP data are based on data from 30 surface stations, CRU data are created by interpolating between all available surface stations in the region, and GPCP data are a combination of satellite and gridded surface measurements. Meteorological station networks tend to have an over representation of low-elevation sites as shown by *Briggs and Cogley* [1996], and since high-elevation sites are often characterized by orographic precipitation enhancement [*Johansson and Chen*, 2003], station averages may underestimate the actual precipitation in the area. We therefore expect gridded data sets of precipitation to have slightly higher values than the SYNOP station averages. Moreover, systematic rain gauge undercatchment due to aerodynamical effects is a well known problem [e.g., *Morris et al.*, 1995], and the correction for such effects in the GPCP data would heighten GPCP precipitation levels further. This manifests itself in the different absolute values in Figure 6, where SYNOP data have consistently the lowest precipitation values while GPCP data have the highest. However, our main aim is not to determine the area-mean precipitation accurately but to investigate the precipitation trends, for which individual station biases against the area mean (e.g., due to topography) are less critical. Between 1983 and 2006, SYNOP data for the BT area showed a 13% increase, GPCP values increased by 10%, and CRU data increased by 5%. The variation in the magnitude of the trends depending on the data set used underlines the statistical insignificance of the precipitation change. We conclude that precipitation levels have increased in the BT region, but that the change is probably even less pronounced than indicated from the uncorrected SYNOP data.

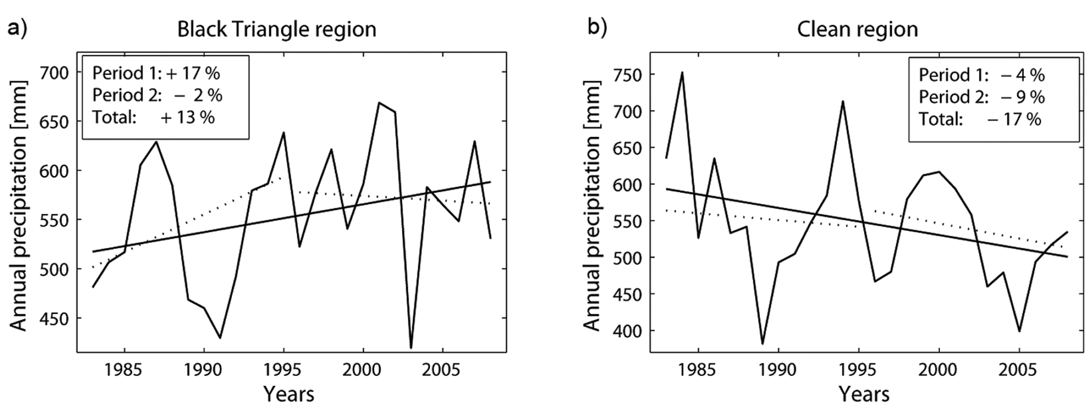

Figure 5. Annual mean precipitation from (a) the Black Triangle area and (b) the clean region, based on SYNOP data. Linear trends for period 1 (1983–1995), period 2 (1995–2008), and the total period (1983–2008) are shown.

Table 2. Trends in Precipitation Variables for the Two Regions[a]

	Annual		Winter		Summer	
	BT	Clean Region	BT	Clean Region	BT	Clean Region
Precipitation amount (mm)	+13%	−17%	+10%	−6%	+11%	+8%
	(+70.8)	(−92.7)	(+11.5)	(−10.0)	(+19.1)	(+6.3)
Heavy precipitation days	+24%	+5%	+48%	+15%	+31%	+14%
	(+1.9)	(+0.5%)	(+0.4)	(+0.3)	(+1.0)	(+0.2)
Light precipitation days	+13%	+56%[b]	+6%	+57%[b]	+22%[c]	+74%[b]
	(+8.6)	(+34.9)	(+1.0)	(+8.6)	(+2.8)	(+10.0)
Cumulus cloud days	+11%[c]	+0%	−2%	+4%	+2%	+3%
	(+20.1)	(+0.1)	(−0.3)	(+0.8)	(+1.6)	(+1.3)
Stratus cloud days	+3%	−6%	+0%	+4%	−7%	−3%
	(+7.2)	(−13.3)	(+0.3)	(+2.2)	(−3.8)	(−1.5)
Rain shower days	+23%[b]	−16%	+12%	−14%	+26%[c]	+3%
	(+23.7)	(−11.3)	(+1.9)	(−2.6)	(+8.4)	(+0.4)
Drizzle days	−17%	−5%	+13%	+2%	−35%	+15%
	(−5.7)	(−2.3)	(+1.8)	(+0.3)	(−1.9)	(+1.4)
Precipitation days	+5%	+13%[c]	−1%	+8%	+8%	+37%[c]
	(+7.7)	(+20.6)	(−0.3)	(+1.6)	(+3.0)	(+11.2)
Precipitation days, N	+37%[c]	+4%	−0%	+38%	−4%	+1%
	(+33.0)	(+2.3)	(−0.0)	(+6.0)	(−1.1)	(+0.1)
Precipitation days, S	+10%	−52%[c]	−44%	−79%	+34%	−15%
	(+2.7)	(−20.6)	(−1.2)	(−5.1)	(+3.3)	(−1.1)
Precipitation days, E	+27%	−16%	+1%	+5%	+36%	−44%
	(+18.9)	(−7.7)	(+0.1)	(+0.4)	(+10.1)	(−3.3)
Precipitation days, W	+2%	−16%	+12%	−7%	+3%	+37%
	(+6.7)	(−65.0)	(+11.1)	(−9.4)	(+2.9)	(+22.3)
Light precipitation days, N	+31%[c]	+47%[c]	+49%	+26%	−6%	+97%[c]
	(+2.6)	(+3.2)	(+1.4)	(+0.4)	(−0.1)	(+1.2)
Light precipitation. days, S	+10%	+73%[b]	−53%	+44%	+45%	+96%[c]
	(+0.4)	(+4.8)	(−0.3)	(+0.5)	(+0.3)	(+1.3)
Light precipitation. days, E	+24%	+89%[b]	+3%	+68%	+59%	+87%
	(+1.8)	(+6.9)	(+0.1)	(+1.2)	(+0.7)	(+1.0)
Light precipitation days, W	+5%	+44%[b]	+5%	+48%[c]	+19%	+66%[b]
	(+2.4)	(+18.3)	(+0.8)	(+5.1)	(+1.8)	(+6.5)

[a]Annual, winter (December through February) and summer (June through August) trends (in percent; trends in absolute values are given in parenthesis) in various SYNOP parameters for the Black Triangle (BT) and clean region (C). Trend for total precipitation is based on annual precipitation sums (mm), while all other variables are based on annual number of events. Postfixes N, S, E and W indicate events occurring during northerly, southerly, easterly and westerly winds, respectively. Values are rounded up to nearest integer.

[b]1% significance.

[c]5% significance.

[27] If the small precipitation increase observed emanates from a causal relationship between pollution levels and precipitation, we would expect to see a precipitation decrease in periods when pollution levels increased. According to *Vestreng et al.* [2007] there was a general increase in sulphur dioxide emissions in Europe from 1945 to around 1980. We therefore compare CRU precipitation trends in the "reduction period" 1983–2006 to a period representative of increasing pollution loads. For this purpose, the years 1947–1970 are chosen to give a period which is of equal length as the reduction period and which ends before the major emission reductions in Europe started. Figure 7 gives annual precipitation sums as well as 5 year running averages between 1901 and 2006 for the BT. While precipitation had a statistically insignificant increase of 5% between 1983 and 2006, precipitation values in the emission increase period increased insignificantly by 12%, contrary to the decrease expected if aerosol changes had a dominating effect on the precipitation changes. However, the large year-to-year variability of the time series implies a high sensitivity of the trend to the exact period chosen, and emission increase periods can be found for which the trend is much weaker or even decreasing. More important than the magnitude of the trend, however, is the general impression that total precipitation does not seem to bear an aerosol signal, with for instance precipitation suppression during the period of highest pollution levels (1970s and 1980s) compared to periods before and after.

6.2. Geographic Variations in Total Precipitation Trends

[28] The above results indicate that total precipitation rates are not notably affected by sulphate concentrations, but to get a more general picture we now look at the spatial distribution of the precipitation trends for all of Europe using the CRU data set. If sulphate concentrations have a strong effect on the precipitation trends, we should see larger precipitation trends in heavily industrialized regions than in clean regions. In Figure 8 we present precipitation trends for the same two periods as above. Figure 8b showing the 1983–2006 period confirms the results already found in the SYNOP data (see Figure 5); a weak decrease in precipitation in the clean regions and an increase in the BT. There is a tendency for the precipitation increase to be strongest in eastern Europe, where emission reductions have been largest, but no opposite tendency can be seen in the 1947–1970 period, when both the coastal clean region and the BT region display precipitation increases (Figure 8a).

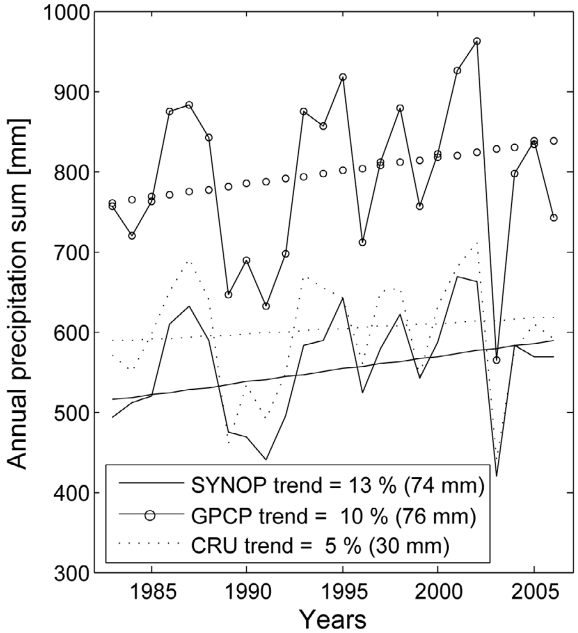

Figure 6. Comparison of 1983–2006 precipitation in the BT area based on SYNOP, CRU, and GPCP data. Linear trends are also shown.

6.3. Trends in Different Precipitation Types

[29] As discussed in section 1, the sign and magnitude of aerosol-precipitation effects may vary depending on the conditions. *Goswami et al.* [2006] studied precipitation trends in India and pointed out that the lack of a significant trend in precipitation amounts most likely was caused by significant but opposite trends in heavy and moderate precipitation, respectively, underlining the need to study these precipitation types separately.

[30] The annual number of days with heavy and light precipitation for both the BT and clean areas is shown in Figures 9a and 9b and trends are given in Table 2. Table 2 shows a large annual trend in the frequency of heavy precipitation events in the BT. Note, however, that annual trends for both the BT and the clean region are statistically insignificant (*p* values 0.07 and 0.81, respectively) as well as very small in absolute values; for example, an increase of less than 2 heavy precipitation days from 1983 to 2008 in the BT. A significant increase of 23% (*p* value 0.002) in frequency of rain showers and of 11% (*p* value 0.017) in observations of cumulus cloud types over the same period (Table 2) may instead indicate that the increase in heavy precipitation in the BT is driven by factors other than aerosol-cloud interactions. For instance, absorbing aerosols (e.g., soot) have been shown to enhance atmospheric stability, thereby suppressing shallow cloud formation over land [*Koren et al.*, 2008]. In our case, a decrease in aerosol concentrations may have caused increased surface heating, enhancing summertime convection and potentially the occurrence of rain showers. However, such an effect would be largest in the period of largest emission reduction, while the number of summertime rain showers only increased in the 1995–2008 period (not shown). Alternatively, changes in land use and urban growth (likely to have been largest after the recovery of the economic breakdown in the area in the late 1980s) have also been shown to affect convective precipitation [*van den Heever and Cotton*, 2007] and may have contributed to the observed trends. Heavy precipitation in the clean region showed only weak changes.

[31] As for light precipitation, Figure 9b reveals that the number of days with light precipitation has increased in both regions. The BT and clean region have increases in the annual number of light precipitation events of 13% (statistically insignificant with *p* value 0.09) and 56% (significant at the 1% level), respectively (see Table 2). To test the sensitivity of these trends to the light precipitation threshold applied, we calculated trends also for events with less than 1, 2, or 3 mm over 12 hours. For both regions the trends weakened when increasing the threshold (see Table 3), possibly indicating a transition from an aerosol-influenced regime for very light precipitation events to a regime of little aerosol influence for more intense precipitation. The light precipitation increases occurred predominantly in the 1983–1995 period (BT, +12%; clean region, +11%) for which sulphur concentrations decreased the most. However, a decrease in light precipitation events of similar magnitude characterized the 1996–2008 period for the BT (−9%) while a small (+2%) increase occurred in the clean region. In both regions, the light precipitation trends are by far the largest in the summertime, but for the clean region the increase is significant also in wintertime.

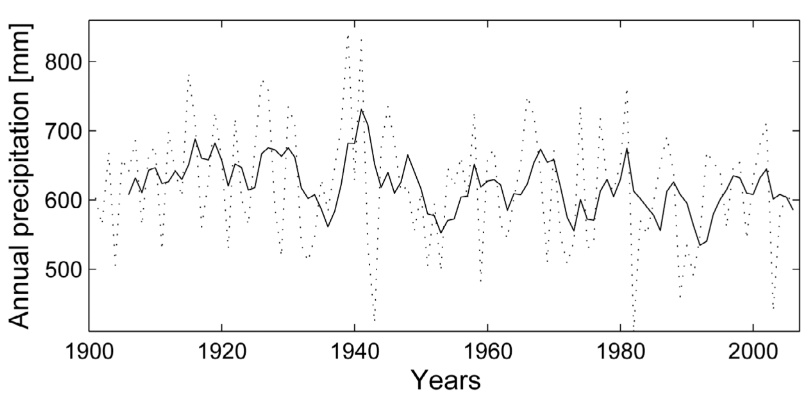

Figure 7. Annual (dotted) and 5 year running averages (solid) of precipitation in the Black Triangle area in the 20th century, based on CRU data.

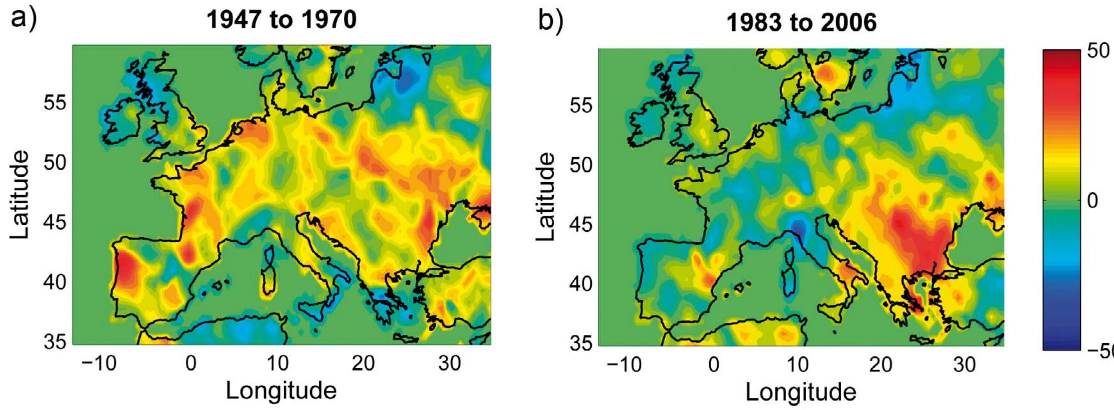

Figure 8. Spatial distribution of trends (% of mean) in monthly precipitation for Europe, based on CRU data, for (a) a period with a mean sulphur emission increase in Europe and (b) a period with a mean emission decrease.

[32] In concordance with these trends we also find for the BT statistically insignificant increases in the frequency of observed drizzle and stratus cloud types between 1983 and 1995 and highly significant decreases in the 1996–2008 period (not shown). An increase in light precipitation in the 1983–1995 period might be a sign of lessened precipitation suppression caused by diminishing aerosol concentrations, but no such explanation can be offered to the stronger trends of the second period. So far, we have shown trends in total precipitation and in particular precipitation forms, and will now look at changes in these variables during specific wind directions and weather types. Such stratifications should help to remove the contributions from other factors, thus facilitating the detection of a potential aerosol effect.

6.4. Dependence on Wind Direction

[33] We hypothesize that along the west coast of Europe, westerly winds are associated with low aerosol concentrations while continental winds from the east contain higher levels of pollutants. In the BT, westerly winds should likewise be associated with cleaner air, whereas easterly and northerly winds have passed over industrialized areas and carry polluted air to the region. To underpin this assumption we show, in Table 4, SO_2 concentrations for different wind directions for one EMEP station in the BT and one EMEP station in the clean region. Wind stratifications were performed using ECMWF reanalysis data (ERA-40 and ERA-Interim). Vector winds from two points (one in the BT area and one in the France area) were extracted for the 850 hPa level to avoid wind directions affected by local topography.

[34] For both stations the concentrations are lowest for westerly winds, but relative variations for various wind directions are larger for the coastal station than for the BT station. The 1983–2008 SO_2 changes (not shown) for the coastal station are largest (a decrease from 9.4 to 1.5 $\mu g/m^3$) for easterly winds while at the BT station the trend is largest (from 17.9 to 1.7 $\mu g/m^3$) for northerly winds; both trends are significant at the 1% level. Smaller variability in the BT than

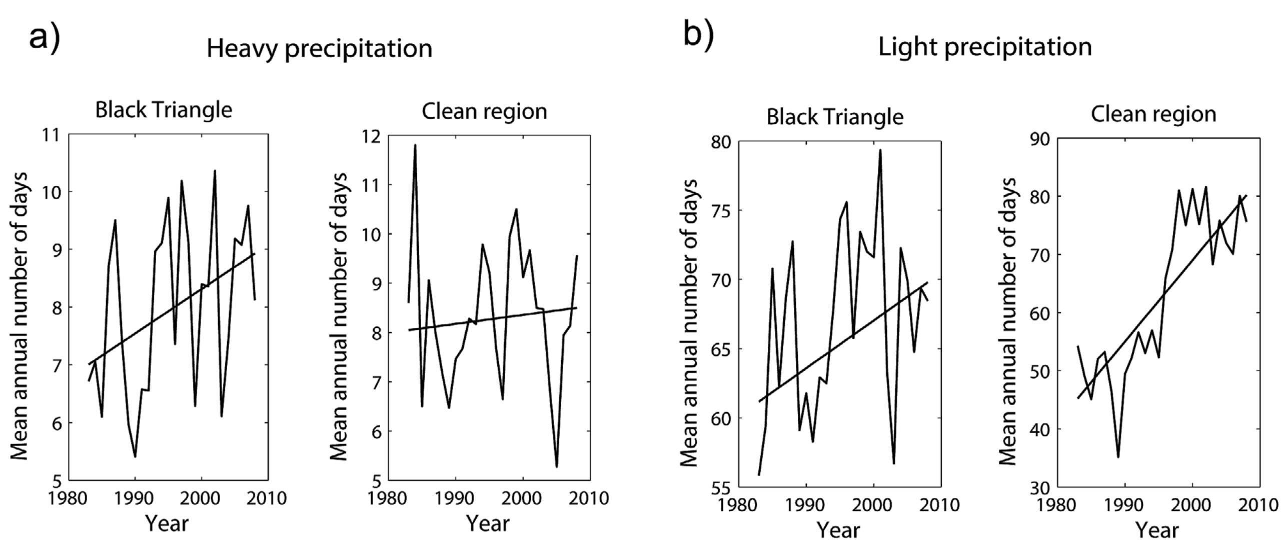

Figure 9. SYNOP data showing the annual number of (a) heavy and (b) light precipitation days between 1983 and 2008; see Table 2 for the magnitudes of the trends.

Table 3. Trends in the Annual Number of Light Precipitation Events in the Black Triangle and Clean Region at Different Light Precipitation Thresholds for the 1983–2008 Period[a]

Precipitation Threshold	Black Triangle (%)	Clean Region (%)
<0.5 mm	13	56
<1.0 mm	10	36
<2.0 mm	7	27
<3.0 mm	6	20
>0 mm	5	13

[a]Originally defined as <0.5 mm over 12 hours.

in the clean region is also found when studying trends in horizontal visibility stratified by wind direction: while the 1983–2008 visibility trend in the BT area for the different wind directions varies only between 14.0 and 14.8 km, the change in the clean region over the same period is +1.7 km for westerly winds and +4.3 km for easterly winds.

[35] To study precipitation trends for different wind directions we have stratified daily precipitation according to the governing wind direction of each day and calculated trends in the number of days with precipitation for each wind direction (see Table 2). Based on the above considerations, the probability of detecting an aerosol signal in precipitation in the clean area should be highest (lowest) for easterly (westerly) winds. However, Table 2 shows a significant trend in the annual number of days with precipitation only for southerly winds in the clean region. In the BT, on the other hand, we find a significant increase of 37% in the annual number of days with precipitation during northerly winds, for which SO_2 concentrations and trends were largest. This increase occurs in the pollution reduction period 1983–1995, while the 1996–2008 period has a weak decrease in precipitation frequency.

[36] As shown in Table 2, a significant 1983–2008 trend in light precipitation frequency appears in the BT also for northerly winds. Recall that no significant change in this parameter was found when considering all wind directions (see Figure 9). Interestingly, the clean region reveals significant increases in light precipitation for all wind directions, but the relative increase is largest for easterly winds for which SO_2 concentrations have declined the most. While the absolute values of these changes are minor, they may bear the signal of an aerosol effect. The 89% increase for easterly winds in the clean region means that for an average station in this area the annual number of light precipitation events during easterly winds increased from 4 to 11 days from 1983 to 2008. However, the increase was larger in the 1996–2008 period than in the 1983–1995 period, for which we would expect the largest aerosol influence on the trend.

6.5. Dependence on Circulation and Weather Type

[37] While we could not find any sign of aerosol influence on total precipitation in the above analysis, changes in atmospheric circulation patterns may constitute an important nonaerosol contribution to variations in surface precipitation amounts. Therefore, a study of changes in the North Atlantic Oscillation (NAO) over the last decades was performed, as the various phases of the NAO may be associated with above-normal or below-normal precipitation levels in different regions of Europe [*Hurrell*, 1995]. The wintertime NAO index shows a decreasing trend between 1983 and 2008, which could possibly induce a trend in precipitation. However, we fail to find any correlation between the NAO index and precipitation sums in the BT area. Indeed, *Niedźwiedź et al.* [2009] found that the NAO index only accounts for 4% of annual precipitation variability in this area, but that other circulation indices (the *C* index) were largely responsible for the observed increase and current decline in the late twentieth century–early twenty-first century precipitation totals in east central Europe, which can be recognized also for the BT in Figure 5a.

[38] As an alternative to the NAO index we use the subjective Hess and Brezowsky weather pattern classification [*Gerstengarbe and Werner*, 2005], which is based on the mean air pressure distribution (sea level and 500 hPa level) over the North Atlantic Ocean and Europe, and which is believed to be particularly representative for eastern and central Europe and thus for the BT. The classification identifies three groups of circulation types (zonal, mixed, and meridional), which are divided into 29 subtypes ("Grosswetterlagen", henceforth abbreviated GWL). Table 5 shows the 29 GWL, their definitions and associated precipitation trends. We apply these weather types to our data to investigate to what extent the observed precipitation trends can be associated with corresponding trends in the frequency of the different GWL. To do this, we first calculate the 1983–2008 mean daily precipitation for winter, spring, summer, and autumn for each of the 29 GWL (referred to later as the "expected" daily precipitation amount). For each GWL we then multiply the given GWL frequency for a given season with that season's expected daily precipitation amount, and the results are then summed for all GWL creating a circulation-based time series from 1983 to 2008 of seasonal precipitation sums. The seasonal expected precipitation sums are then summed to create annual time series of precipitation amounts. Year-to-year variations and trends in this estimated precipitation time series would then only be induced by circulation changes in terms of the varying frequencies of the different weather types. Since we have used mean precipitation amounts for the various GWL types, aerosols would not have an influence on this trend estimate. In contrast, the original precipitation data also reflect the influence of aerosols, so that any aerosol signal should appear as a residual of the two trend estimates. In summary, the GWL classification is an effective way to reduce the strong influence of synoptic variability on the trend estimates, thus allowing a secondary influence by aerosols, if present, to stand out more clearly in the residuals.

Table 4. Mean 1983–2008 Concentration of SO_2 in the Air at Two EMEP Stations and Station SO_2Concentration at Different Wind Directions

	Mean ($\mu g/m^3$)	N ($\mu g/m^3$)	S ($\mu g/m^3$)	E ($\mu g/m^3$)	W ($\mu g/m^3$)
Coastal station[a] (La Hague, France)	2.4	2.6	2.6	4.2	1.7
Black Triangle station[b] (Svratouch, Czech Republic)	5.3	7.3	5.1	5.4	4.9

[a]Coastal station La Hague, France can be seen as a green dot near the "F" in Figure 1.

[b]BT station Svratouch, Czech Republic can be seen as a green dot in the BT square in Figure 1.

Table 5. Overview of the 29 Hess and Brezowsky Weather Subtypes[a] (GWL)

	Name	Definition	Mean Frequency (d/yr)	R Deviation (mm)	SO_2 Deviation ($\mu g/m^3$)	GWL Trend	R Trend (mm)	SO_2 Trend ($\mu g/m^3$)
1	WA	Anticyclonic Westerly	20	−0.54	1.45	−16.70	−0.27	−12.46
2	WZ	Cyclonic Westerly	62	0.50	−1.34	−4.50	−0.28	−8.64
3	WS	South-shifted Westerly	6	0.56	−2.33	−6.20	0.95	−3.74
4	WW	Maritime Westerly (block eastern Europe)	6	0.34	0.40	0.00	0.22	−10.80
5	SWA	Anticyclonic South-Westerly	14	−0.86	−0.52	−8.30	0.01	−8.79
6	SWZ	Cyclonic South-Westerly	16	0.22	−2.71	15.60	0.03	−4.00
7	NWA	Anticyclonic North-Westerly	11	−0.27	1.72	−8.30	0.90	−11.81
8	NWZ	Cyclonic North-Westerly	20	1.22	−1.65	7.10	0.07	−8.57
9	HM	High over central Europe	19	−1.18	3.42	−5.70	−0.07	−11.75
10	BM	Zonal ridge across central Europe	45	−0.67	1.61	11.10	0.28	−12.77
11	TM	Low (cut-off) over central Europe	8	1.58	1.91	−5.80	1.35	−11.65
12	NA	Anticyclonic Northerly	2	0.41	1.83	0.00	−0.33	−14.70
13	NZ	Cyclonic Northerly	7	0.91	2.09	0.00	0.19	−14.34
14	HNA	Iceland high, ridge central Europe	7	−0.78	−0.25	0.00	−0.03	−9.29
15	HNZ	Iceland high, through central Europe	6	0.57	−0.85	−8.30	−1.11	−4.16
16	HB	High over the British Isles	13	−0.65	2.58	−7.10	−0.40	−15.68
17	TRM	Through over central Europe	24	1.04	−1.68	16.20	0.46	−8.93
18	NEA	Anticyclonic North-Easterly	3	−0.17	1.12	0.00	0.43	−3.38
19	NEZ	Cyclonic North-Easterly	3	1.99	−1.05	0.00	−2.82	−4.20
20	HFA	Scandinavian high, ridge central Europe	9	−0.99	1.00	0.00	0.73	−9.38
21	HFZ	Scandinavian high, through central Europe	4	−0.41	3.73	0.00	−0.51	−9.92
22	HNFA	High Scandinavia-Iceland, ridge central Europe	6	−1.10	−1.09	0.00	−0.15	−6.06
23	HNFZ	High Scandinavia-Iceland, through central Europe	5	−0.21	−0.03	3.10	0.36	−4.06
24	SEA	Anticyclonic South-Easterly	8	−1.22	2.20	5.00	0.05	−9.14
25	SEZ	Cyclonic South-Easterly	3	−0.10	−1.10	1.50	0.19	−4.95
26	SA	Cyclonic Southerly	5	−1.28	2.07	0.00	−0.03	−12.95
27	SZ	Anticyclonic Southerly	1	−0.71	−0.97	0.00	0.23	−4.42
28	TB	Low over the British Isles	7	0.20	−2.14	−5.00	−0.11	−5.71
29	TRW	Through over western Europe	20	0.38	−1.57	4.50	0.36	−7.14

[a]Columns show for each GWL the frequency of occurrence (in days per year), the daily precipitation deviation based on SYNOP BT data (mean daily precipitation for days with the given GWL only minus mean daily precipitation based on all days, which will give a positive number for GWLs associated with above-normal daily precipitation amounts), a similar deviation for SO_2 concentration at Svratouch in the Czech Republic, 1983–2008 linear trend in the annual frequency of the given GWL, 1983–2008 linear trend in the daily precipitation amounts for days with the given GWL only, and a similar linear trend for SO_2 concentration.

[39] Observed (dotted line) and GWL-estimated (solid line) annual precipitation sums are shown for the SYNOP data in Figure 10a. While the observed precipitation between 1983 and 2008 increased by 13%, the GWL-estimated precipitation increased by 11% over the same period, indicating that variations in weather types and thus atmospheric circulation patterns may be connected to the majority of the trend in total precipitation. It is important to note here that the changing GWL frequency would have lead to an increase in visibility of 0.9 km over the 1983–2008 period, whereas the observed increase over this period was 15 km (not shown). Thus, visibility trends were almost entirely driven by the strong reduction of aerosol loads and cannot be explained by changes in the meteorological conditions, in contrast to the precipitation changes. Also shown in Figure 10b is the residual between the observed and the GWL-estimated precipitation, which should bear the signatures of all other effects than the changing frequencies of GWL. While year-to-year variations, presumably caused by factors other than aerosol changes, remain in the residual plot, there is no visible trend corresponding to the large changes in aerosol concentrations. Similar results were found for GPCP and CRU data (not shown).

[40] Both average aerosol loadings and precipitation formation mechanisms (and thus precipitation types) depend on the synoptic conditions and will vary between different GWL classes. Therefore, another approach to investigate potential aerosol effects is to study precipitation trends for specific GWL classes, for which these characteristics are more homogeneous than for the total data set. For instance, a potential precipitation signal from emission reductions over the past decades may be particularly clear if we focus on precipitation that fell during weather types associated with large trends in aerosol concentrations. Using daily SO_2 values from the EMEP station Svratouch in Czech Republic and stratifying by GWL type, we find among the five most frequent GWL the largest relative SO_2 trend of −112% (from 1983 to 2008, compared to 1983 values) for the weather type "anticyclonic westerly" (WA). This weather type is associated with stagnant conditions and does not often yield precipitation, but when it does it is likely to be strongly affected by the local aerosol concentration. But while a large effect from decreased precipitation suppression by the diminishing aerosol concentrations would induce an increase in daily precipitation amounts associated with the WA weather type, we find a statistically insignificant *decrease* of 0.27 mm in the daily mean precipitation between 1983 and 2008 (see Table 5). Only one GWL class was associated with a significant change in daily precipitation, but this weather type did not display a particularly large decrease in SO_2 concentrations (see GWL type 20 in Table 5). We do not find any specific GWL for which a clear aerosol signal can be identified in trends of daily mean precipitation amounts.

7. Discussion

[41] We find that mean sulphate concentrations at 12 EMEP stations in central Europe were 72% lower in 2008

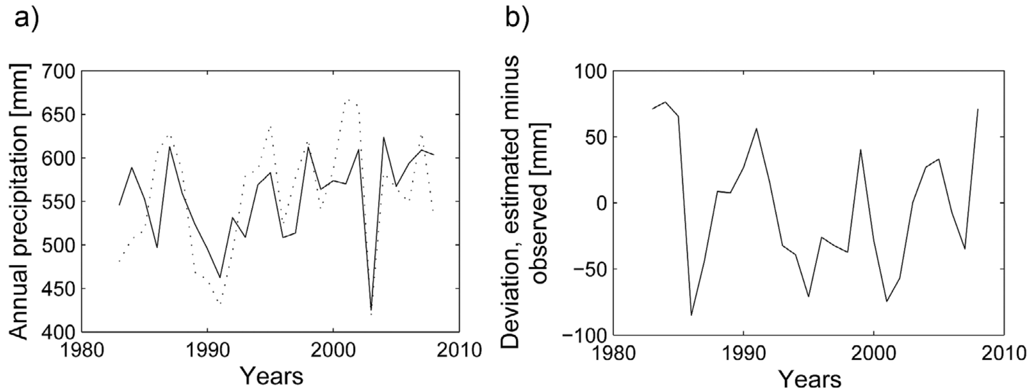

Figure 10. (a) Observed (dotted) and GWL-estimated (solid) annual precipitation for the BT and (b) anomaly (estimated minus observed) precipitation, based on the 29 weather types and the SYNOP precipitation data.

than in 1983. The BT was chosen as our area of focus, being the most polluted region of Europe in the 1980s. Horizontal visibility measurements during nonfoggy days in the BT show for the same period an increase in visibility from 11 to 27 km, confirming the presence of a substantial decrease in aerosol concentrations in this specific area, and indicating that the strong emission changes had a large impact on the radiation budget. However, an impact on precipitation is less clear.

7.1. Total Precipitation

[42] In the above analyses, we find no aerosol signal in trends of total precipitation in the period of emission reductions in Europe. Specifically, changes in total precipitation between 1983 and 2008 were not statistically significant for either the BT (+13%) or the clean region (−17%). Moreover, precipitation for a period of increasing pollution, namely 1947–1970, showed a statistically insignificant increase of 12% for the BT. Whereas levels and trends in aerosol concentrations have been vastly different between polluted and clean areas of Europe, spatial trend maps based on gridded precipitation data showed no consistent difference in precipitation trends between such areas. For a similar period (1971–2005), *Bokwa* [2009] compared total precipitation from rural and urban stations in Poland but found no signal from decreasing pollution levels in the precipitation time series, consistent with our findings.

[43] Precipitation from cold shallow clouds (such as the stratiform clouds typically observed in wintertime in both the BT and clean region) may be subject to invigoration during high ice nuclei concentrations [*Wallace and Hobbs*, 1977]. As soot particles have shown a potential to function as ice nuclei [*Gorbunov et al.*, 2001], and as it is reasonable to assume that decreasing pollution in general and modernization of power plants will have lead to decreased soot emissions [*Wehner et al.*, 1999], this will have contributed to lowering precipitation amounts from cold shallow clouds. However, we find no clear wintertime trends in total precipitation: while the clean region saw a weak decrease in wintertime precipitation levels, the BT area saw a similar increase.

7.2. Convective Precipitation

[44] We searched for signs of aerosol invigoration of precipitation from convective clouds with warm cloud bases by studying summertime trends in heavy precipitation events as well as events with rain showers. If this effect is important in Europe, the theoretical basis, as summarized in section 1, implies decreased trends in the above parameters during the past 30 years of emission reductions. However, the BT had a significant (and the clean region a small but insignificant) increase in summertime rain showers and both areas had insignificant increases in heavy precipitation events. Cloud bases even in summertime may not have been sufficiently warm (~15°C [*Rosenfeld et al.*, 2008]) to support the invigoration effect, and studies have found precipitation suppression in cooler convective clouds [e.g., *Khain et al.*, 2001] which may instead have induce the increases observed. Alternatively, low relative humidities in this continental region may have caused efficient evaporation, which could also lead to convective precipitation suppression [*Khain*, 2009]. However, a closer look at the summertime BT rain showers showed that the significant increase took place in the 1995–2008 period, which had only minor pollution reductions. Consequently, we do not believe that aerosols have had a large effect on heavy precipitation in these areas.

7.3. Light Precipitation

[45] In the 1983–2008 period of strong emission reductions, the clean region saw a significant increase in light precipitation of 56% while the BT experienced a smaller and statistically insignificant increase of 13%. The significant trend in the clean region is particularly interesting as the total precipitation amount was observed to decrease over the same period. Conceivably, higher susceptibility to aerosol changes in this area of lower background concentrations [see, e.g., *Andreae et al.*, 2004; *van den Heever and Cotton*, 2007] may have allowed an aerosol-precipitation signal to appear more clearly here. As light precipitation in wintertime is likely to be associated with cold shallow clouds that may be subject to opposite aerosol effects, we looked at summer trends separately and found even stronger increases (significant trends of 22% and 74% for the BT and clean region, respectively). Moreover, increasing the threshold for our definition of light precipitation weakened the trends, possibly indication a transition from an aerosol-influenced regime for the lightest precipitation types to a regime of lower

precipitation susceptibility for the more intense precipitation types.

[46] A similar relationship between total and light precipitation trends was found by *Qian et al.* [2009] for Eastern China, where sulphur emissions increased by about 25 Tg/yr in the period 1956–2005 [see, also, *Liu et al.*, 2010]. The same period was marked by increase/decrease in total precipitation depending on area, but light precipitation decreased in their whole study region, presumably as a result of precipitation suppression by aerosols. By comparison, Europe experienced a 40 Tg/yr *decrease* in sulphur dioxide emissions over the past three decades [*Vestreng et al.*, 2007], and while we found that total precipitation increased (decreased) in the BT (clean region), light precipitation *increased* in both regions.

[47] For the BT area, trends in both the annual number of days with precipitation and the frequency of light precipitation events were larger (and statistically significant) for northerly winds, for which trends in SO_2 were observed to be largest, than for all other wind directions. For the clean region, light precipitation during both continental easterly and maritime westerly winds was associated with significantly increased light precipitation frequency. The increase was largest for easterly winds, but it should be noted that the change was larger in the 1996–2008 period than in the 1983–1995 period, for which most of the pollution reductions occurred.

[48] From the above, we see that the increase in light precipitation is compatible with a decreasing aerosol effect on precipitation during the last decades.

7.4. Alternative Causes of the Observed Precipitation Trends

[49] An investigation of aerosol effects on precipitation must be accompanied by an account of the influence of other factors on precipitation variations. Global warming increases the atmosphere's saturation vapor pressure and may therefore intensify the hydrological cycle and thus precipitation [*Ramanathan et al.*, 2001]. *Hulme et al.* [1998] found a global precipitation sensitivity of 1.5–2.5% per degree global warming for the 1900–1996 period, with exact sensitivities varying from region to region. This means that the observed precipitation trends may in part have been induced by warming effects, but are not likely to have been caused by atmospheric warming alone. Changes in atmospheric circulation patterns, on the other hand, are a major contributor to precipitation variations, and we therefore studied the variability in Hess and Brezowsky weather types to elucidate underlying aerosol trends. A strong coherence between variations in weather types and precipitation was revealed. We created an estimated precipitation time series for the BT based on changing frequency of weather types, and found that this circulation-induced precipitation trend was of similar magnitude (+11%) as the observed precipitation trend (+13%), presumably because the frequency of weather types associated with above-normal precipitation has increased. Similarly, no aerosol signal could be found when studying precipitation variations separately within GWL types, for which other sources of variability would be filtered out. This means that the observed changes in precipitation can mostly be explained by changes in the frequency of particular weather situations, without invoking a mechanism requiring the presence of aerosols.

[50] A note of caution is in place, as we cannot rule out that precipitation was influenced indirectly by aerosols via changing the frequency of particular weather types. *Ménégoz et al.* [2010] studied the connection between wintertime aerosol loads and weather regimes in Europe, and found for instance that aerosol patterns induced by a positive NAO episode could destabilize and reduce the persistence of a subsequent negative NAO episode. However, this would not modify precipitation mechanisms via microphysical cloud properties as studied in this paper.

8. Conclusions

[51] In the present article we investigate the effect of aerosols on precipitation in the Black Triangle. After 1989, economic effects following the political changes in the region, as well as subsequent pollution mitigation measures, induced dramatic improvements of air quality in the area. If aerosols have had an effect on precipitation trends in Europe, such a signal may be most pronounced the BT region. The nonlinearity of aerosol-precipitation interactions combined with large natural variability in precipitation time series complicates the detection of very small signals. Therefore, our ambition has not been to provide conclusive evidence on any particular precipitation modification mechanism, but rather to explore whether a potential aerosol influence is so large that it is possible to find an aerosol signal in multidecadal precipitation data.

[52] The analyses presented above showed the following:

[53] 1. SO_2 emissions decreased by 90% in central Europe, measured sulfate concentrations decreased by 72%, and mean horizontal visibility in the BT improved from 11 to 27 km between 1983 and 2008.

[54] 2. Total precipitation showed no sign of an aerosol signal, and a circulation-based precipitation estimate could explain 11% of the 13% increase in precipitation amounts between 1983 and 2008 in the BT.

[55] 3. No aerosol signal was found in heavy precipitation.

[56] 4. A possible aerosol signal was visible in light precipitation, which increased in both the BT and clean region, particularly for wind directions associated with the largest reductions in pollution levels. Trends were largest for the smallest light precipitation thresholds, and largest in summertime; possibly due to cancelling effects of ice nuclei in shallow winter clouds.

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References

Adler, R. F., et al. (2003), The version 2 Global Precipitation Climatology Project (GPCP) monthly precipitation analysis (1979–present), *J. Hydrometeorol.*, *4*, 1147–1167.

Aitken, J. (1880), On dust, fogs, and clouds, *Nature*, *23*, 195–197, doi:10.1038/023195d0.

Albrecht, B. A. (1989), Aerosols, cloud microphysics, and fractional cloudiness, *Science*, *245*, 1227–1230, doi:10.1126/science.245.4923.1227.

Alpert, P., P. Kishcha, Y. J. Kaufman, and R. Schwarzbard (2005), Global dimming or local dimming?: Effect of urbanization on sunlight availability, *Geophys. Res. Lett.*, *32*, L17802, doi:10.1029/2005GL023320.

Alpert, P., N. Halfon, and Z. Levin (2008), Does air pollution really suppress precipitation in Israel?, *J. Appl. Meteorol.*, *47*, 933–943, doi:10.1175/2007JAMC1803.1.

Andreae, M. O. (2009), Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions, *Atmos. Chem. Phys.*, *9*, 543–556, doi:10.5194/acp-9-543-2009.

Andreae, M. O., D. Rosenfeld, P. Artaxo, A. A. Costa, G. P. Frank, K. M. Longo, and M. A. F. Silva-Dias (2004), Smoking rain clouds over the Amazon, *Science*, *303*, 1337–1342, doi:10.1126/science.1092779.

Ardo, J., N. Lambert, V. Henzlik, and B. N. Rock (1997), Satellite-based estimations of coniferous forest cover changes: Krusne Hory, Czech Republic 1972–1989, *Ambio*, *26*, 158–166.

Ayers, G. P. (2005), Air pollution and climate change: Has air pollution suppressed rainfall over Australia?, *Clean Air Environ. Qual.*, *39*, 51–57.

Ayers, G. P., and Z. Levin (2009), Air pollution and precipitation, in *Clouds in the Perturbed Climate System*, edited by J. Heintzenberg and R. J. Charlson, pp. 369–400, MIT Press, Cambridge, Mass.

Bell, T. L., D. Rosenfeld, K. M. Kim, J. M. Yoo, M. I. Lee, and M. Hahnenberger (2008), Midweek increase in U.S. summer rain and storm heights suggests air pollution invigorates rainstorms, *J. Geophys. Res.*, *113*, D02209, doi:10.1029/2007JD008623.

Blas, M., M. Sobik, and R. Twarowski (2008), Changes of cloud water chemical composition in the Western Sudety Mountains, Poland, *Atmos. Res.*, *87*, 224–231, doi:10.1016/j.atmosres.2007.11.004.

Bokwa, A. (2009), Effects of air pollution on precipitation in Kraków (Cracow), Poland in the years 1971–2005, *Theor. Appl. Climatol.*, *101*, 289–302, doi:10.1007/s00704-009-0209-7.

Briggs, P. R., and J. G. Cogley (1996), Topographic bias in mesoscale precipitation networks, *J. Clim.*, *9*, 205–218

Dusek, U., et al. (2006), Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science*, *2*, 1375–1378, doi:10.1126/science.1125261.

Fan, J., T. Yuan, J. M. Comstock, S. Ghan, A. Khain, L. R. Leung, Z. Li, V. J. Martins, and M. Ovchinnikov (2009), Dominant role by vertical wind shear in regulating aerosol effects on deep convective clouds, *J. Geophys. Res.*, *114*, D22206, doi:10.1029/2009JD012352.

Gerstengarbe, F. W., and P. C. Werner (2005), Katalog der Grosswetterlagen Europas (1881–2004) (in German), *Rep. 100*, 153 pp., Potsdam Inst. Klimafolgenforschung, Potsdam, Germany.

Givati, A., and D. Rosenfeld (2004), Quantifying precipitation suppression due to air pollution, *J. Appl. Meteorol.*, *43*, 1038–1056.

Gorbunov, B., A. Baklanow, N. Kakutkina, H. L. Windsor, and R. Toumi (2001), Ice nucleation on soot particles, *J. Atmos. Sci.*, *32*, 199–215, doi:10.1016/S0021-8502(00)00077-X.

Goswami, B. N., V. Venugopal, D. Sengupta, M. S. Madhusoodanan, and P. K. Xavier (2006), Increasing trend of extreme rain events over India in a warming environment, *Science*, *314*, 1442–1445, doi:10.1126/science.1132027.

Gunn, R., and B. B. Phillips (1957), An experimental investigation of the effect of air pollution on the initiation of rain, *J. Atmos. Sci.*, *14*, 272–280.

Halfon, N., Z. Levin, and P. Alpert (2009), Temporal rainfall fluctuations in Israel and their possible link to urban and air pollution effects, *Environ. Res. Lett.*, *4*, 025001, doi:10.1088/1748-9326/4/2/02500.

Hulme, M., T. J. Osborn, and T. C. Johns (1998), Precipitation sensitivity to global warming: Comparison of observations with HadCM2 simulations, *Geophys. Res. Lett.*, *25*, 3379–3382.

Hurrell, J. W. (1995), Decadal trends in the North Atlantic Oscillation and relationships to regional temperature and precipitation, *Science*, *269*, 676–679, doi:10.1126/science.269.5224.676.

Ishizaka, Y., and M. Adhikari (2003), Composition of cloud condensation nuclei, *J. Geophys. Res.*, *108*(D4), 4138, doi:10.1029/2002JD002085.

Jirak, I. L., and W. R. Cotton (2006), Effect of air pollution on precipitation along the Front Range of the Rocky Mountains, *J. Appl. Meteorol.*, *45*, 236–245.

Johansson, B., and D. Chen (2003), The influence of wind and topography on precipitation distribution in Sweden: Statistical analysis and modeling, *Int. J. Climatol.*, *23*, 1523–1535, doi:10.1002/joc.951.

Johnson, D. B. (1982), The role of giant and ultragiant aerosol particles in warm rain initiation, *J. Atmos. Sci.*, *39*, 448–460.

Khain, A. P. (2009), Notes on state-of-the-art investigations of aerosol effects on precipitation: A critical review, *Environ. Res. Lett.*, *4*, 015004, doi:10.1088/1748-9326/4/1/015004.

Khain, A. P., D. Rosenfeld, and A. Pokrovsky (2001), Simulating convective clouds with sustained supercooled liquid water down to −37.5°C using a spectral microphysics model, *Geophys. Res. Lett.*, *38*, 3887–3890, doi:10.1029/2000GL012662.

Koren, I., J. V. Martins, L. A. Remer, and H. Afargan (2008), Smoke invigoration versus inhibition of clouds over the Amazon, *Science*, *321*, 946–949, doi:10.1126/science.1159185.

Krüger, O., and H. Graßl (2002), The indirect aerosol effect over Europe, *Geophys. Res. Lett.*, *29*(19), 1925, doi:10.1029/2001GL014081.

Kulmala, M., A. Laaksonen, P. Korhonen, T. Vesala, T. Ahonen, and J. C. Barrett (1993), The effect of atmospheric nitric acid vapor on cloud condensation nucleus activation, *J. Geophys. Res*, *98*, 22949–22958.

Levin, Z., and W. R. Cotton (Eds.) (2008), *Aerosol Pollution Impact on Precipitation*: *A Scientific Review*, 386 pp., Springer, New York.

Liu, B., M. Xu, and M. Henderson (2010), Where have all the showers gone? Regional declines in light precipitation events in China, 1960–2000, *Int. J. Climatol.*, doi:10.1002/joc.2144.

Ménégoz, M., V. Guemas, D. S. y Melia, and A. Voldoire (2010), Winter interactions between aerosols and weather regimes in the North Atlantic European region, *J. Geophys. Res.*, *115*, D09201, doi:10.1029/2009JD012480.

Morris, C. J. G., M. Nunez, and S. Montes (1995), A field study of orographic precipitation anomalies during high velocity winds using an aerofoil rain gauge, *Theor. Appl. Climatol.*, *51*, 1434–4483, doi:10.1007/BF00867440.

New, M., M. Hulme, and P. Jones (2000), Representing twentieth-century space-time climate variability, part II: Development of 1901–1996 monthly grids of terrestrial surface climate, *J. Clim.*, *13*, 2217–2238.

Niedźwiedź, T., R. Twardosz, and A. Walanus (2009), Long-term variability of precipitation series in east central Europe in relation to circulation patterns, *Theor. Appl. Climatol.*, *98*, 337–350, doi:10.1007/s00704-009-0122-0.

Qian, Y., D. Gong, J. Fan, L. R. Leung, R. Bennartz, D. Chen, and W. Wang (2009), Heavy pollution suppresses light rain in China: Observations and modeling, *J. Geophys. Res.*, *114*, D00K02, doi:10.1029/2008JD011575.

Ramanathan, V., P. J. Crutzen, J. T. Kiehl, and D. Rosenfeld (2001), Aerosols, climate, and the hydrological cycle, *Science*, *294*, 2119–2124, doi:10.1126/science.1064034.

Rosenfeld, D. (2000), Suppression of rain and snow by urban and industrial air pollution, *Science*, *287*, 1793–1796, doi:10.1126/science.287.5459.1793.

Rosenfeld, D., U. Lohman, G. B. Raga, C. D. O'Dowd, M. Kulmala, S. Fuzzi, A. Reissell, and M. O. Andreae (2008), Flood or drought: How do aerosols affect precipitation?, *Science*, *321*, 1309, doi:10.1126/science.1160606.

Schultz, D. M., S. Mikkonen, A. Laaksonen, and M. B. Richman (2007), Weekly precipitation cycles? Lack of evidence from United States surface stations, *Geophys. Res. Lett.*, *34*, L22815, doi:10.1029/2007GL031889.

Shepherd, J. M. (2005), A review of current investigations of urban-induced rainfall and recommendations for the future, *Earth Interact.*, *9*, 1–27.

Stephens, B., and G. Feingold (2009), Untangling aerosol effects on clouds and precipitation in a buffered system, *Nature*, *461*, 607–613, doi:10.1038/nature08281.

Teller, A., and Z. Levin (2006), The effects of aerosols on precipitation and dimensions of subtropical clouds: A sensitivity study using a numerical cloud model, *Atmos. Chem. Phys.*, *6*, 67–80, doi:10.5194/acp-6-67-2006.

Thorsheim, P. (2006), *Inventing Pollution*: *Coal*, *Smoke*, *and Culture in Britain Since 1800*, Ohio Univ. Press, Athens, Ohio.

Twomey, S. (1977), The influence of pollution on the shortwave albedo of clouds, *J. Atmos. Sci.*, *34*, 1149–1152.

van Aardenne, J. A., F. J. Dentener, J. G. J. Olivier, C. G. M. Klein Goldewijk, and J. Lelieveld (2001), A 1° × 1° resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990, *Global Biogeochem. Cycles*, *15*, 909–928, doi:10.1029/2000GB001265.

van den Heever, S. C., and W. R. Cotton (2007), Urban aerosol impacts on downwind convective storms, *J. Appl. Meteorol.*, *46*, 828–850, doi:10.1175/jam2492.1.

Vautard, R., P. Yiou, and G. J. van Oldenborgh (2009), Decline of fog, mist, and haze in Europe over the past 30 years, *Nat. Geosci.*, *2*, 115–119, doi:10.1038/ngeo414.

Vestreng, V., G. Myhre, H. Fagerli, S. Reis, and L. Tarrason (2007), Twenty-five years of continuous sulphur dioxide emission reduction in Europe, *Atmos. Chem. Phys.*, *7*, 3663–3681, doi:10.5194/acp-7-3663-2007.

Wallace, J. M., and P. V. Hobbs (1977), *Atmospheric Science An Introductory Survey*, 467 pp., Academic, San Diego, Calif.

Wehner, B., T. C. Bond, W. Birmili, J. Heintzenberg, A. Wiedensohler, and R. J. Charlson (1999), Climate-relevant particulate emission characteristics of a coal-fired heating plant, *Environ. Sci. Technol.*, *33*, 3881–3886, doi:10.1021/es981052f.

Wild, M. (2009), Global dimming and brightening: A review, *J. Geophys. Res.*, *114*, D00D16, doi:10.1029/2008JD011470.

Yue, S., P. Pilon, and G. Cavadias (2002), Power of the Mann–Kendall and Spearman's rho tests for detecting monotonic trends in hydrological series, *J. Hydrol.*, *259*, 254–271, doi:10.1016/S0022-1694(01)00594-7.

J. E. Kristjánsson and C. W. Stjern, Department of Geosciences, University of Oslo, PO Box 1047, Blindern, N-0316, Oslo, Norway. (camilla.stjern@geo.uio.no)

A. Stohl, Norwegian Institute for Air Research, PO Box 100, N-2027, Kjeller, Norway.