Influence of pollution on cloud reflectance

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[1] After the collapse of the East Bloc in 1989, the political and economic changes resulted in significant reductions of industrial activities and thus atmospheric pollution that modified cloud reflectance over and in the lee of the main European emission sources. This impact during a two-decade transition (1981–1999) of atmospheric pollution in Europe, in particular in East Germany and Poland, was studied on the basis of emission data, measured aerosol concentrations, and satellite observations of cloud reflectance. In these main European emission areas the high degree of air pollution generally enhanced variability of cloud reflectance during the 1980s. The variability was strongest for the early 1980s. A distinct influence of increased particle number density and increased black carbon content as well as secondary aerosol formation is detected. Toward the late 1990s, both the radius effect and the absorption effect, as the two components of the so-called first indirect aerosol effect, have declined because of reduced particulate matter and sulphur dioxide emissions. The results indicate a pronounced influence of stability on the indirect aerosol effect over Central Europe. The analyzed frequency distributions of cloud reflectance show characteristics that are in line with the theory of radiative transfer. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* indirect aerosol effect, air pollution, black carbon, sulphur dioxide, cloud albedo, Black Triangle

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1. Introduction

[2] *Twomey* [1977] discussed the influence of air pollution on the shortwave albedo of clouds. He postulated that air pollution acts to increase the reflectance (albedo) of clouds by an increase of cloud droplet concentration. On the other hand, an increase of the absorption coefficient of aerosols alone would lead to a decrease of the reflectance. In the past this so-called first indirect aerosol effect, sometimes called the Twomey effect, was of interest in many mostly theoretical investigations. The third full assessment report of the *Intergovernmental Panel on Climate Change* (*IPCC*) [2001] indicated that the mean global radiative forcing caused by the indirect aerosol effect is negative and could reach -2 W/m^2.

[3] Lately, we investigated this indirect aerosol effect on a regional scale over parts of Europe on the basis of long-term satellite measurements by advanced very high resolution radiometers (AVHRR) [*Krüger and Graßl*, 2002]. The main result was a reduction in mean cloud reflectance between the late 1980s and the late 1990s due to an air pollution decrease in both Western and Central Europe.

[4] In the present article we focus on the variability of cloud reflectance due to anthropogenic activity for the same area. Our hypothesis was as follows: In polluted areas, cloud reflectance should vary more strongly as reflectance is higher on average and varies depending on the direction of flow.

[5] The paper concentrates on the impact of atmospheric pollution on cloud reflectance variability during the episode of strongest emissions in parts of Europe. In order to investigate the reflectance transition patterns, we focus on the industrialized regions of Poland and Germany, which were part of the earlier so-called Black Triangle of Europe. The most important emission centers were located in the heavily industrialized Krakow-Katowice region, in southern Poland, and the other southern part of the former German

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Democratic Republic (East Germany) including the cities of Halle, Leipzig, Bitterfeld, and Cottbus. These areas, which strongly influenced the cloud reflectance over Europe as shown by *Krüger and Graßl* [2002], are described in more detail. The study combines remotely sensed cloud properties and emission data to trace the impact of changed air pollution on cloud optical properties over a large part of Europe.

[6] Since knowledge of the temporal and spatial variability of the aerosol concentration is a basic requirement for the interpretation of cloud optical properties, we first give an overview about pollution in East Germany and Poland within section 2, while the variability of cloud reflectance derived from satellite measurements is described in section 3. The results are then discussed in section 4.

2. Air Pollution in Poland and East Germany

[7] During the last two decades, enormous amounts of air pollutants, both particulate matter and the aerosol precursor gases sulphur dioxide (SO_2) and nitrogen oxides (NO_x) have been emitted mostly into the atmospheric boundary layer, but during the winter and nighttime they have also into the free troposphere from high power plant stacks over Europe.

[8] Before 1989 the economical buildup of Central and Eastern European countries (CEEC) was governed by a continuous growth of heavy industry branches like metallurgy, shipyards, military production complexes, and power plants. These activities demanded a large energy supply and were connected with wasteful energy use mainly from hard coal or lignite. As a result the maximum atmospheric pollution load was observed during the 1980s.

[9] From 1989 onward, radical changes in politics and the economy reshaped all industry branches, but especially those responsible for discharge of atmospheric pollutants. Since the policies for a transition undertaken by individual countries depended on complex sociological and economic conditions, they differed and still differ in speed and efficiency. In general, sudden changes took place in eastern Germany, but they were of a gradual nature in other CEEC. In the case of Poland, the largest country within the CEEC, the transition incorporated both the transfer of large, mostly inefficient companies into the market economy and the restoration of small, technology-oriented market activities. Such a transition for large industry branches like metallurgy or coal mining required complex multistage implementations. The growth of more energy-efficient market activities since 1990 resulted in the reduction of atmospheric pollution.

[10] Extremely high loads of particulate and gaseous atmospheric pollution in Europe originated from specific industry areas in Poland and eastern Germany. Among the pollutants with a potentially strong impact on the indirect aerosol effect are particulate matter (PM), sulphur dioxide (SO_2), nitrogen oxides (NO_x), and ammonia (NH_3). Emission of anthropogenic particulate matter, often called primary particulate matter (PPM), stems predominantly from public electric power, district heating, industrial combustion, commercial and residential combustion, road transport, emissions from industrial processes, and agriculture. The source category of inorganic PPM has as a primary contributor solid fuel combustion at large point sources, e.g., the cement, iron, and steel industries. Organic compounds and black carbon (BC) mainly originate from mobile sources, small combustion, and petroleum extraction and refining.

[11] Emission inventories have been compiled not only for total PPM [*Berdowski et al.*, 1997] but also for black carbon (BC) [*Cooke and Wilson*, 1996; *Liousse et al.*, 1996], which is mainly elemental and to a minor part organic carbon, for heavy metals, e.g., lead and cadmium, and for persistent organic pollutants [*Umweltbundesamt*, 1994a]. While the location of main source regions, especially point sources, is well known, information about the interannual variability of emissions in Europe during the last 20 years is largely lacking. *Cooke and Wilson* [1996] estimated the BC emission of Western Europe to be $\sim$1 Tg C yr^{-1}.

[12] For Germany, annual total PPM emissions were estimated by *Umweltbundesamt* [1994b]. While for the western part the annual emissions amounted to 0.4–0.6 Tg yr^{-1} during 1985–1989, the values for the much smaller eastern part were 4 times higher, reaching 2.2–2.5 Tg yr^{-1} PPM. The emissions in East Germany, the former German Democratic Republic (GDR), originated from large lignite power plants, which accounted for approximately 50% of total emission. More than 300 large combustion plants contributed to the PPM emissions, and stacks higher than 100 m were nearly ubiquitous. The emission data show a clearly decreasing trend for western and eastern parts of Germany since the middle and late 1980s, respectively.

[13] Besides direct emission of PPM, the formation of secondary inorganic aerosol (SIA) and biogenic secondary organic aerosol (BSOA) also contribute to atmospheric aerosol mass. While BSOA is seen as a minor fraction of lower than 5% of the total particulate matter, SIA is the most important contributor in Central Europe with 50–70% [*European Monitoring and Evaluation Programme* (*EMEP*), 2000]. The emissions of the precursor gases SO_2, NO_x, and NH_3 contributing to SIA are the best known of all anthropogenic emissions since the United Nations Economic Commission for Europe initiated the European Monitoring and Evaluation Programme (EMEP) already in 1979. The EMEP emission database includes annual totals of the above species since 1980. The major part of this database contains official emissions submitted by the European countries [*Vestreng and Storen*, 2000]. The highest precursor gas emissions occurred in the 1980s. The emission estimates published by the Meteorological Synthesizing Centre West (MSC-W) of EMEP [see *Vestreng and Storen*, 2000] amount to 53 Tg SO_2 in 1985 for all of Europe. During this time, important contributors from Central Europe were Germany and Poland. In addition, emissions originating from the United Kingdom, France, Netherlands, Czech Republic, and Ukraine contributed considerably to air pollution in Germany and Poland because of long-range transport. Within the period between 1985 and 1989 in most of the European countries the SO_2 emissions decreased slightly by $\sim$10%.

[14] In 1989 the situation changed [*Mylona*, 1999]. Almost immediately after the collapse of the East Bloc, a significant decrease with respect to atmospheric pollution was observed in East Germany, while the changes in other

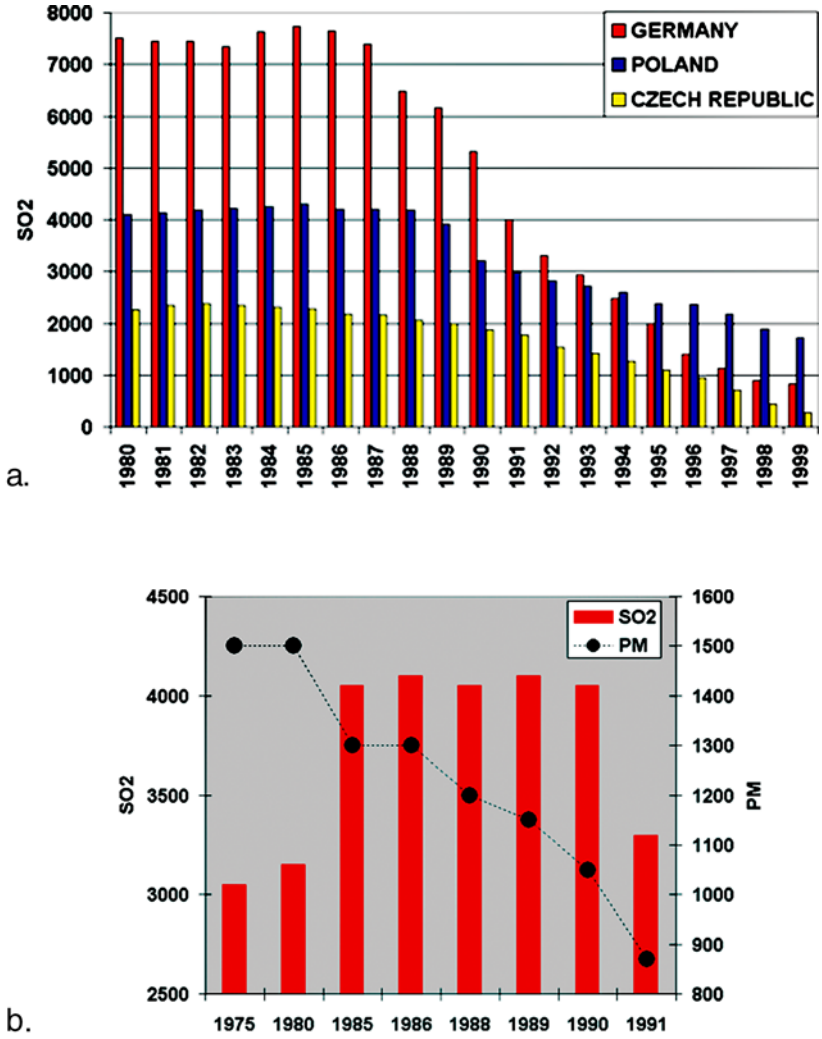

Figure 1. (a) Annual sulphur dioxide (SO_2) emissions from Germany (including the former GDR), Poland, and the Czech Republic, in kilotons for the period 1980–1999 according to the EMEP database [*Mylona*, 1999]. (b) Annual emissions of particulate matter (PM) and sulphur dioxide (SO_2) for power plants in the area of the former GDR, in kilotons. Data are from the German Environmental Agency [*Umweltbundesamt*, 1994b]. (c and d) Model-predicted monthly average sulphur dioxide concentration in ambient air over Europe in 1980 for January (Figure 1c) and November (Figure 1d) (units are $\mu g\ S\ m^{-3}$).

CEEC, including Poland, were more gradual (see Figure 1a). The strong decrease in atmospheric pollution in the former German Democratic Republic (GDR) was related to massive reduction of high stack emissions due to radically closed or modernized industry. In Poland the first signs of pollution decrease were seen for low-level emissions, mostly for residential combustion, which was followed by gradual reduction of emissions from public electric power plants, district heating, and industrial combustion. Significant reduction in atmospheric pollution in both countries was enacted for high stack emissions from industry by deploying exhaust filtration and desulphurization or (additionally in Germany) denoxification systems or modernization of technology by industry. The resulting strong decrease of the sulphur emission in Europe by approximately 50%, from 49 Tg in 1988 to 26 Tg in 1998, has been reported by EMEP [*Vestreng and Storen*, 2000]. In contrast to SO_2 the emission database reflects only a weak decrease of 10–20% for NO_x and NH_3 since 1990 for all of Europe.

[15] The decrease of emissions was confirmed by in situ measurements in main source areas of pollution. For example, in the city of Krakow a significant reduction of atmospheric pollution was observed from 1994 to 1998, when the emission of particulate matter and gases from industry and electric power plants decreased by ~50% [*Turzański and Paula-Wilga*, 1999]. In the Katowice area (Upper Silesia), where the transformation of industry was massive, the concentration of particulate matter with a diameter smaller than 10 μm (PM_{10}) decreased 2.3 times from 1989 to 1996 and turned into a steady annual decrease by a factor of 1.3–1.4 from 1993 to 1996 [*Pastuszka*, 1997]. In the north of Poland, in the urbanized region of Gdynia, close to the Gulf of Gdańsk, known for shipyard and transportation activities, a reduction of particulate matter

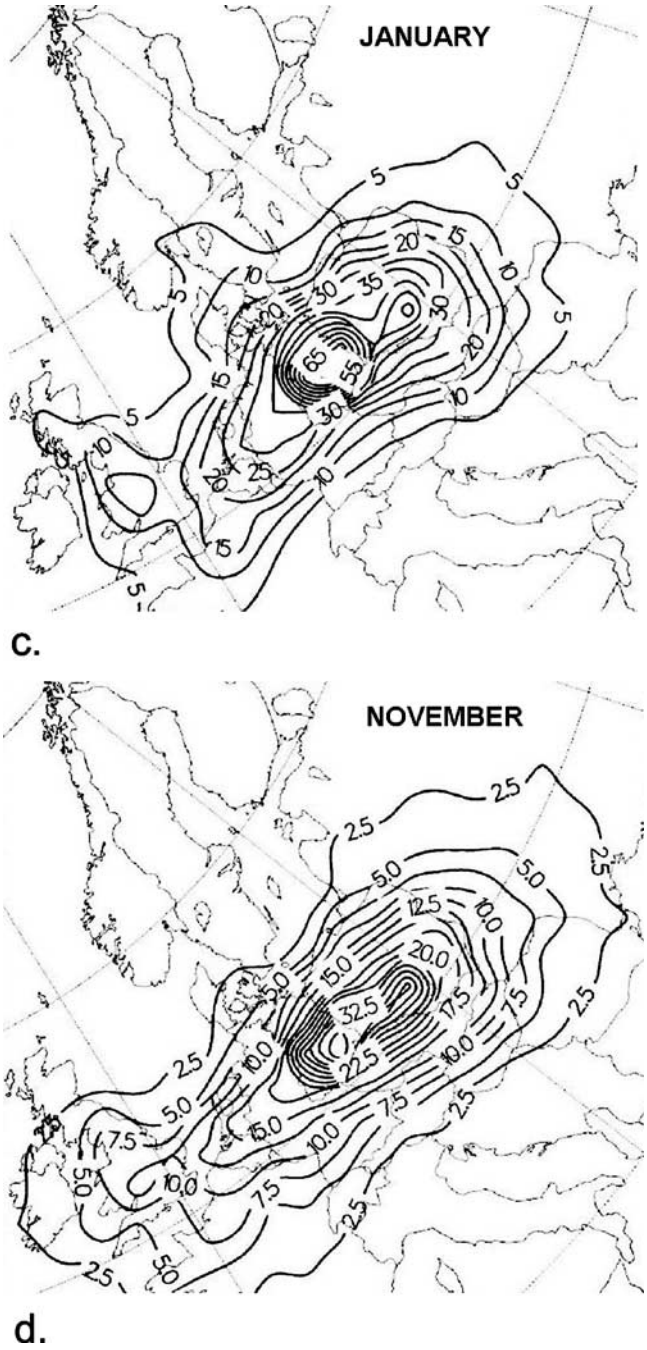

Figure 1. (continued)

in air has been observed since 1972. There the total particulate matter (PM) load into air showed an annual reduction of 15%. Since 1991, about a 30% reduction of total annual PM was observed over the northwest region of Poland (J. Woroń, EMEP representative for Poland, Institute for Meteorology and Water Management, Gdynia, Poland, personal communication, 2000). A reduction twice as high in similar maritime regions in northwest Poland indicates that the pollution that previously originated over East Germany has significantly impacted PM concentrations over northwest Poland, as southwesterly winds dominate in that region.

2.1. Krakow and Katowice

[16] In Poland, especially in the urbanized Krakow-Katowice region, both gaseous and aerosol pollution, but in particular sulphate and carbonaceous aerosols, stabilized at a very high level. The Krakow-Katowice area was regarded as having one of the highest atmospheric pollution levels of all cities in Poland. In 1997, for example, the city of Krakow was rated as the third highest in particulate matter (PM) concentration, with an annual emission of 10,900 t. For the same year the annual emission of polluting gases was 103,400 t, including 26,900 t for SO_2 alone [*Turzański and Paula-Wilga*, 1999]. Within the city area, extending 30 km west to east and 15 km north to south, are located 30 industry-related major atmospheric pollution point sources. These include the largest Polish metallurgy plant (Huta Sendzimira), situated ~15 km to the east, and the large coal-fired electric power plant Skawina, located only 15 km southwest of the city center.

[17] In Krakow, a significant decrease in the atmospheric pollution trend has been noticed after ~2 years of transition. The gradual modernization of industry and, in particular, a significant reduction of coal combustion plants including metallurgy industries, accompanied by the growth of a new, more energy-efficient market economy, resulted in an extended transition period with respect to atmospheric pollution. In addition, the emissions from individual households showed a decrease since 1991 [*Turzański and Paula-Wilga*, 1999]. That process has been stimulated by the reduction of coal combustion, as more oil and natural gas are used nowadays for heating. This caused a major reduction of near-surface atmospheric pollution and has led to improved air quality in the Krakow-Katowice urbanized areas.

[18] In the Krakow area the air quality is strongly influenced by calm conditions that are frequent over the low-lying river valleys of the Wisła, Rudawa, Wilga, Białucha, and Dłubnia, which are located within the city of Krakow. These conditions favor accumulation of air pollution and the formation of smog, especially during periods with temperature inversions, existing 36–42% of the time [*Turzański and Paula-Wilga*, 1999].

[19] The city of Katowice, located only ~50 km from Krakow in the center of Upper Silesia, is the largest highly industrialized area in Poland. Within that area, hundreds of large and small heavy industry plants are located. The trend in atmospheric pollution is difficult to estimate. The averaged data on the concentrations of particulate matter with a diameter lower than 2.5 μm ($PM_{2.5}$) and PM_{10} were given as 138 and 185 μg m^{-3}, respectively, for 1990 [*Pastuszka*, 1997]. The values have decreased to ~60 μg m^{-3} toward the end of the 1990s [*Pastuszka*, 1997; *Turzański and Paula-Wilga*, 1999].

[20] During the early 1980s a slightly increasing tendency of the total load of particulate matter in the atmospheric boundary layer was replaced by a slight decrease starting in 1986. From 1991 to 1993 an extremely fast PM concentration reduction of 60 μg m^{-3} was observed [*Pastuszka*, 1997; *Turzański and Paula-Wilga*, 1999]. Afterward, the reductions of both particulate and gaseous pollution continued, reaching 60% from 1994 to 1998 [*Turzański and Paula-*

Wilga, 1999]. It has to be pointed out that these decreasing emissions are in disagreement with a publication by *Berdowski et al.* [1997] that speaks about a slight increase of PM_{10} and $PM_{2.5}$ emissions for all of Poland, from 93,000 to 100,000 t per year for the 1990–1993 period.

2.2. Halle, Leipzig, and Cottbus

[21] During the 1980s, another area of very high atmospheric pollution was East Germany. There a large part of the total gaseous and particulate emission was emitted from elevated stacks of lignite combustion plants, accounting for ~50% of the PM emissions. For gaseous emissions the contribution of power plants was even higher, for example, reaching more than 75% for SO_2. Since the emission height of many point sources lay above 100 m, the formation of secondary aerosols has been favored.

[22] The main emission centers of particulate and gaseous pollutants in the GDR were located around the cities of Halle, Leipzig, and Cottbus. In 1989, more than 200 of 460 point sources were situated in and around these cities.

[23] The PM emissions in the former GDR decreased nearly continuously from the beginning of the 1980s [*Umweltbundesamt*, 1994b], when they had reached ~2.6 Tg. The decline amounted to ~1 Tg for the source categories of power plants and industry and ~0.7 Tg only for the power plants (see Figure 1b). However, there was an interruption of the continuous downward trend in the PM emissions during the 1980s. Because of a considerable cut in the supply of oil from the Soviet Union in 1982 a renaissance of lignite took place. The consequence was a slight increase of less than 0.1 Tg PM emission until 1986. After 1986 the decrease of the PM emissions was the most pronounced, and it was due to filtration of PM in the large stacks.

[24] The SO_2 emissions of the power plants show a different behavior (see Figure 1b). In the beginning of the 1980s the emissions were ~3.1 Tg yr^{-1}, climbing to ~4.1 Tg yr^{-1} [*Umweltbundesamt*, 1994b]. This was a clear exception as compared to the slight decreases during the 1980s in western Central Europe.

[25] The strong contribution of elevated point sources around Halle, Leipzig, and Cottbus resulted in pronounced spatial differences in PM concentrations in air. For example, in Cottbus, enormous amounts of PM were emitted by the industrial complex "Gaskombinat Schwarze Pumpe." The emission of individual point sources there reached at least 10,000 t yr^{-1}. In the region Halle-Leipzig, quite a number of point sources were located at the chemical plants "Chemie AG Bitterfeld-Wolfen," but in Schkopau (Halle), "Chemische Werke Buna" was also a major emission area.

[26] Consequently, the air concentrations of pollutants showed a pronounced maximum in the former GDR accompanied by spatial heterogeneity in Central Europe in general. Our model estimates (for model details, see *Eliassen and Saltbones* [1983] and *Krüger and Tuovinen* [1997]), based on emission data for 1980 [*Meinl et al.*, 1989], confirm a strong monthly variability due to distinct removal by dry and wet deposition processes. Also, increased air concentrations occur predominantly eastward of the major power plant emissions in the former GDR and in Poland (see Figures 1c and 1d). A few years later, during the late 1980s, the measured PM and SO_2 concentration peaks show values even higher than 150 μg m^{-3} around Halle, Leipzig, and Cottbus, while the concentrations in rural areas were much lower, by ~50 μg m^{-3} [*Umweltbundesamt*, 1994b].

[27] In the beginning of the 1990s, PM and SO_2 concentrations rapidly declined because of massive emission reductions. The annual mean concentration values for SO_2 were lower by ~50% in rural areas and by ~70% in the cities as compared to peak values in 1985. The PM concentrations showed a similar strong decrease, which again, as for SO_2, was dependent on the region [*Umweltbundesamt*, 1994b].

[28] We expected that such strong emission reductions in both PM and SO_2 should have major consequences for optical properties of clouds. Therefore we investigated changes in cloud reflectance and its variability as continuously observed from space since the early 1980s over the former GDR and the surrounding areas of Central Europe.

3. Variability of Cloud Reflectance

[29] In order to study the variability of cloud reflectance we evaluated the reprocessed global National Oceanic and Atmospheric Administration (NOAA)/NASA Pathfinder data set from the advanced very high resolution radiometer (AVHRR) on board the afternoon NOAA series satellites (NOAA 7, 9, 11, and 14) [*James and Kalluri*, 1994]. The full five-channel AVHRR multispectral information was used for the detection of different cloud types. For cloud classification we applied the NOAA/National Environmental Satellite Data and Information Service algorithm [*Stowe et al.*, 1991].

[30] Since the optical properties of lower clouds are expected to be influenced more strongly by pollution than those of high clouds, our investigations were restricted to low- and medium-level clouds. After cloud classification the near-infrared channel 2 reflectance was further evaluated as it is nearly insensitive to Rayleigh scattering and has no strong contribution from aerosols above the low clouds.

[31] The idea was to compare time periods of distinct emissions of PM and SO_2. Therefore two episodes during the 1980s, from 1981 to 1984 and from 1985 and 1989, and one during the 1990s, from 1996 to 1999 (denoted as 8184, 8589, and 9699, respectively), were chosen.

[32] Because the height of the atmospheric boundary layer and solar radiation intensity itself have an influence on the concentration of pollutants, two seasons, namely, the winter months (January, February, November, and December, denoted as JFND8184, JFND8589, and JFND9699) and the summer months (May, June, July, and August, denoted as MJJA8184, MJJA8589, and MJJA9699) were evaluated separately.

[33] Since we have found the indirect aerosol effect near the strong emission sources of PM and SO_2 [*Krüger and Graßl*, 2002], the area of interest was again a large part of Central Europe including smaller parts of Western and Eastern Europe (longitude −5°W to 31°E, latitude 48°–60°N). There the influence from the former GDR and Poland, regions of extremely high emissions of primary particles as well as precursor gases during the 1980s, is dominant.

[34] Now we investigated the variability of cloud reflectance in more detail over the well-known areas of strong air

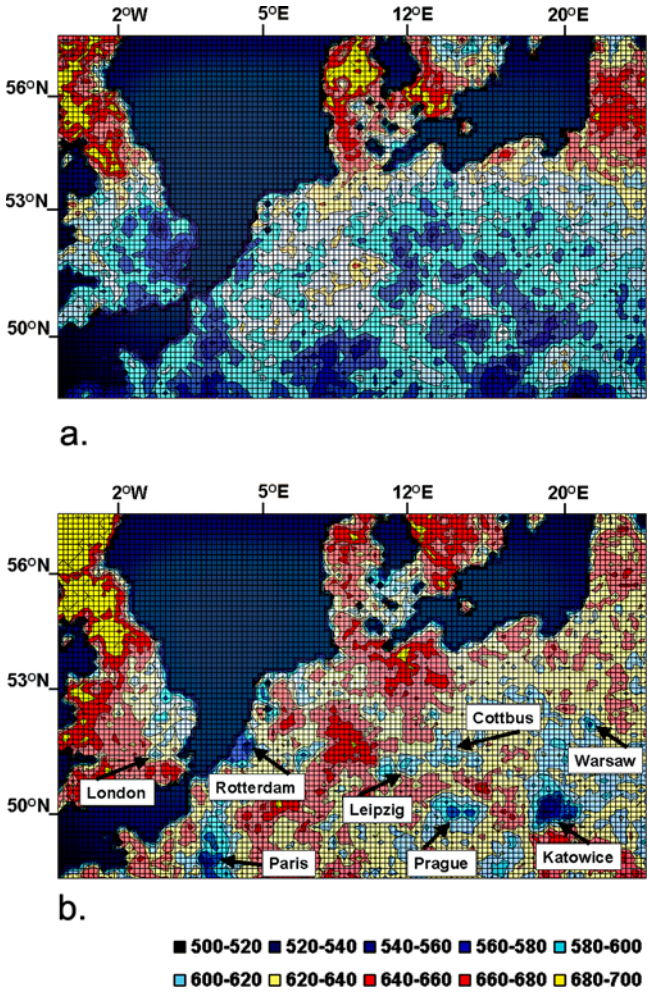

Figure 2. Mean cloud reflectance of low- and medium-level clouds derived from AVHRR channel 2 data over parts of Europe, given in thousands. Data are shown from 5.0°W to 24.0°E longitude and 48.5° to 57.5°N latitude for the winter periods (a) November and December and (b) January and February from 1985 to 1989 (excluding data from 1987). The grid size is 0.25° longitude and 0.125° latitude. The area covers parts of the United Kingdom, Denmark, Sweden, France, Benelux, Germany, Poland, Czech Republic, the Baltic countries, and Ukraine. The North Sea and the Baltic Sea are in black. Strongly polluted areas are indicated.

pollution in Europe where an increased variability had been detected during winter in source regions [*Krüger and Graßl*, 2002]. These areas, in addition, were accompanied by much lower mean reflectance. However, the spatial heterogeneity of the wintertime cloud reflectance over Europe is also remarkable. There is a clear negative gradient of cloud reflectance from Great Britain, Scandinavia, and the former Soviet Union to densely populated industrial regions of Europe, with up to 10% lowered values over strongly polluted regions (see Figures 2a and 2b). The areas of minimum mean cloud reflectance with values even lower than 58% for JFND8589 are located around London, Paris, Rotterdam, Leipzig, Prague, Katowice, Krakow, and Warsaw. The reflectance decrease from "remote" clouds to clouds over source regions is due to the impact of black carbon (BC) emissions originating mainly from power plants, small combustion, and mobile sources. In source regions the single-scattering albedo of water clouds is therefore lower.

[35] Since BC particles are hydrophobic immediately after emission and chemically inert [*Crutzen et al.*, 1984], the lower reflectance of water clouds can be explained by the high amount of absorbing particles in source regions, which favors the physical transformation of black carbon into a hydrophilic form through coagulation with, for example, sulphate or nitrate particles. The condensation process close to source regions is based on internally mixed aerosol with the consequence of an increased portion of BC inside cloud droplets. An additional explanation for the significantly increased absorption might be that low clouds in source regions contain a higher portion of BC particles in the interstitial aerosol. Since the absorption of solar radiation in clouds is thus increased, we call this process the absorption part of the first indirect aerosol effect.

[36] Besides the feature of spatial heterogeneity, there are pronounced differences in the mean reflectance of the total area for the individual episodes. As compared to the periods 1981–1984 and 1996–1999, the mean reflectances for winter and summer are generally increased during the period 1985–1989 (Table 1), which points to the obvious connection to peak values of precursor emissions during the late 1980s. Since strongly increased formation of secondary aerosols took place then, we attribute the increase of reflectance to the radius effect as part of the first indirect aerosol effect [*Krüger and Graßl*, 2002]. The characteristics are quite pronounced even after subdivision into the main emission areas in Germany and Poland, which are the regions around Halle-Leipzig-Cottbus and Krakow-Katowice.

[37] In contrast to mean reflectance the trend of variability shows a different behavior. There is a continuous decrease of variability from JFND8184 to JFND9699. The tendency since the beginning of the 1980s is in good agreement with the continuous reduction of total PM emission in Europe. On the basis of this finding, we argue that emissions of PM are a major factor influencing the variability of cloud reflectance.

[38] In general, the variability of cloud reflectance is smaller for clouds in remote regions, e.g., over the west

Table 1. Mean Cloud Reflectance Derived From AVHRR Channel 2 Data Over Parts of Europe (−5.0° to 24.0° Longitude and 48.5°–57.5°N Latitude) and the Areas Centered Around Halle, Leipzig, and Cottbus (10.0°–15.3° Longitude and 50.8°–52.3°N Latitude) and Around Krakow and Katowice (16.5°–22.3° Longitude and 49.5°–51.5°N Latitude) for the Winter Period January, February, November, and December From 1981 to 1984, 1985 to 1989 (Excluding Data From 1987 Because of Incomplete Data), and 1996 to 1999[a]

	1981–1984	1985–1989	1996–1999
Total Area	60.8 (18.3)	61.5 (17.5)	58.7 (17.4)
Halle-Leipzig	58.5 (20.3)	60.7 (17.7)	58.4 (17.1)
Krakow-Katowice	58.8 (19.3)	59.9 (18.2)	57.1 (18.1)

[a]Mean cloud reflectance is given in percent. The standard deviation is given in parentheses.

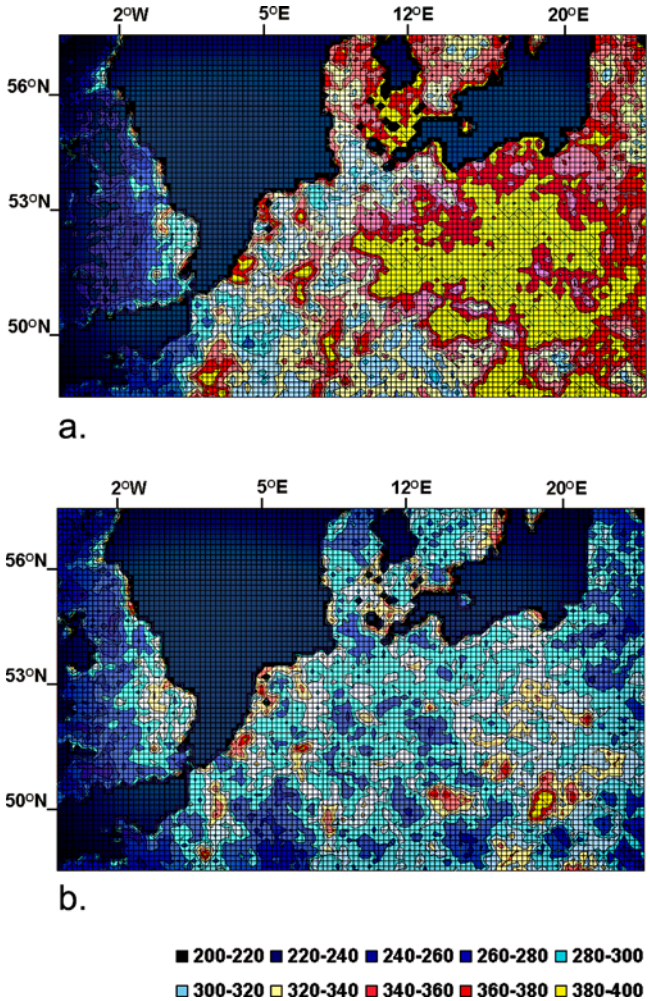

Figure 3. Mean coefficient of variation of cloud reflectance derived from AVHRR channel 2 data over parts of Europe, given in thousands; area is as in Figure 2, but for the four months January, February, November, and December from (a) 1981 to 1984 and (b) 1996 to 1999.

United Kingdom. However, over London and the industrial areas near the British east coast the variability already increases slightly.

[39] Over the European continent, cloud reflectance variability shows highest values in the source regions of PM mainly in the eastern part of our test area. It is obvious that the strongest variability occurred during JFND8184 in Germany and Poland (Figure 3a). The impressive plume of high variability over nearly all regions of the former GDR and Poland corresponds well with the higher degree of pollution extending mainly eastward from the major source regions (see Figures 1c and 1d).

[40] For JFND8589 this feature already declined [see *Krüger and Graßl*, 2002]. During JFND9699 (Figure 3b), only a slightly increased variability, which is limited to the well-known emission regions of PM, is still to be seen. For the Halle-Leipzig-Cottbus and the Kracow-Katowice areas, the hot spots of pollution in Germany and Poland, the general tendency toward lower cloud reflectance is well confirmed. However, as compared to Krakow-Katowice, the Halle-Leipzig and Cottbus regions show a much stronger decrease in variability from the late 1980s to the late 1990s, in agreement with the stronger emission reduction in eastern Germany. In both areas of the former GDR the disappearance of high variability was accompanied by a pronounced change in PM concentrations. Hence our above hypothesis concerning a general dependency of cloud reflectance on the PM concentration in the boundary layer is well supported.

[41] More characteristics of cloud reflectance variability can be found by analyzing the frequency distributions of cloud reflectance. Also, as Figure 2 indicates, a further subdivision of the episodes is useful for analyzing the influence of increased concentrations of pollutants. Therefore we divided the data into episodes of two months, November–December (ND) and January–February (JF). Especially for the low planetary boundary layer heights in full winter, some remarkable details about the aerosol influence on cloud optical characteristics become visible.

[42] We started to look again at the mean reflectance in our area. The intention was to eventually detect characteristics of the indirect aerosol effect for different cloud optical thickness during early and late winter. However, for this question, the change of mean reflectance for JF and ND shows no tendency and therefore does not allow straightforward conclusions.

[43] However, if we compare the change for the individual episodes JF and ND with the emissions of SO_2 and PM, interesting similarities become visible (see Table 2). We see a remarkable dependence of the JF reflectance on SO_2 emissions. The maximum value of 63.1% for JF8589 was reached during the time of strongest SO_2 emissions. For the two other episodes, JF8184 and JF9699, the reflectance was more than 2.5% lower. On the other hand, the reduction of the PM emissions from 1981 to 1999 is accompanied by an even stronger decrease in reflectance from 62.4% for ND8184 to 58.6% for the ND9699 episode. These results already show that a high amount of primary particles during ND8184 as well as enhanced formation of secondary aerosols during JF8589 were leading to an increased radius effect.

[44] A closer look into the variability of cloud reflectance confirms that the strong emission of PM is the main reason for the high variability during JF8184 as well as ND8184. A dependency of variability for the JF episodes on enhanced formation of secondary aerosols during 8589 is not seen. This indicates the presence of at least one additional process influencing the variability during JF beside the radius effect.

Table 2. Mean Cloud Reflectance Derived From AVHRR Channel 2 Data Over Parts of Europe (−5.0° to 24.0° Longitude and 48.5° to 57.5°N Latitude) for the Winter Periods ND, JF, and JFND and for Summer Period MJJA From 1981 to 1984, 1985 to 1989 (Excluding Data From 1987), and 1996 to 1999[a]

	1981–1984	1985–1989	1996–1999
ND	62.4 (19.0)	59.8 (18.1)	58.1 (18.7)
JF	59.4 (17.6)	63.4 (16.8)	59.2 (16.1)
JFND	60.8 (18.3)	61.5 (17.5)	58.7 (17.4)
MJJA	64.9 (12.1)	65.7 (12.5)	63.6 (11.3)

[a]Mean cloud reflectance is given in percent. The standard deviation is given in parentheses. ND, November and December; JF, January and February; JFND, January, February, November, and December; MJJA, May, June, July, and August.

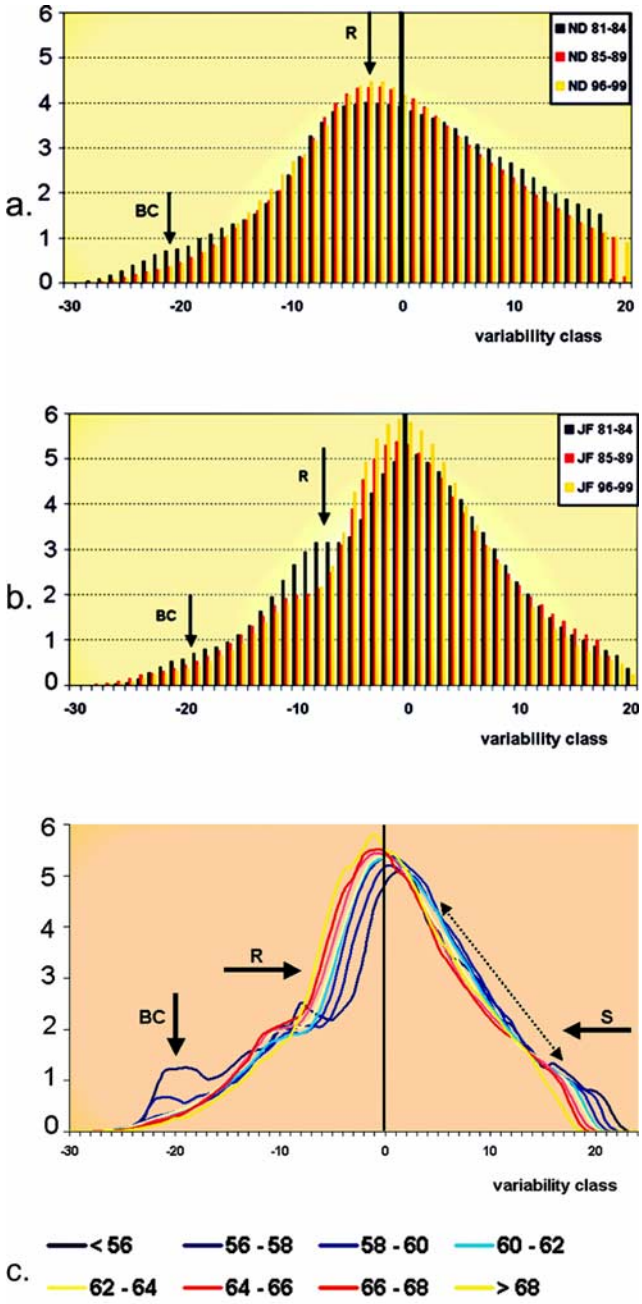

Figure 4. (a and b) Frequency distribution of the deviation from mean cloud reflectance that is shown in Table 2 (in percent per variability class interval of 2% width) for November and December (ND) (Figure 4a) and January and February (JF) (Figure 4b) during 4-year periods (1981–1984, 1985–1989, and 1996–1999), excluding 1987. The black carbon peak (BC) and the radius effect (R) are indicated. (c) Frequency distribution of the deviation from mean cloud reflectance for JF8589 as depicted in Table 2 (in percent per variability class interval of 2% width). Colors refer to different cloud reflectance classes for the episode JFND8589 [see *Krüger and Graßl*, 2002] in percent (source regions, blue; more remote, red and yellow, as in Figure 2). The main characteristics of the first indirect aerosol effect are indicated (R, radius effect; S, saturation effect through aerosol absorption). The black carbon peak (BC) is due to the frequency distribution in source regions of air pollution.

The additional process responsible is the absorption effect described above, which can strongly reduce the cloud reflectance for sufficiently optically thick clouds.

[45] The frequency distributions in Figures 4a–4c show more details regarding the different indirect aerosol effects. The distributions are generally much broader in ND as compared to JF. The shape for ND (Figure 4a) is positively skewed, which already indicates the modification of the normal distribution by pollution. The frequency distribution is broadest during ND8184, pointing to a major impact of PM emissions on the variability. The maximum in the negative branch during ND can be well explained by the radius effect, which rather increases the reflectance of optically thinner clouds, i.e., clouds with lower reflectance. However, the increased frequency for optically thick clouds in ND8184, seen in the positive branch, is also a clear indication of an increased cloud optical depth.

[46] During JF the influence of the radius effect becomes more evident. This is seen from the frequency distribution (Figure 4b) at the range of −20% deviation from the mean. Since the reflectance at this point is ∼40%, the optical thickness should be comparably low at ∼20. For layer clouds of this reflectance class, radiative transfer calculations give the most pronounced increase in cloud reflectance because of the radius effect [*Graßl*, 1978]. In Figure 4b at −20% reflectance the radius effect is seen in a clear shift to higher values, which modifies the shape of the distribution considerably.

[47] Further confirmation of our interpretation can be found by looking at the frequency distributions for areas of different degree of local pollution, i.e., differently lowered mean JFND reflectance (Figure 4c). As discussed above, the spatial heterogeneity of the mean reflectance is considered as an indicator for the BC content in the clouds (see Figure 2). The lower mean reflectance in main pollution areas in Figure 2 can be explained by a higher BC content, i.e., the absorption effect. Therefore source regions of pollution and more remote regions can be identified by subdivision into classes of different mean reflectance.

[48] If we compare now the frequency distributions for these classes of different mean JFND reflectance [see *Krüger and Graßl*, 2002], more details of the indirect aerosol effect become visible (Figure 4c). First, again for optically thinner clouds starting at about −20%, there is a clear shift toward higher reflectance for polluted areas, while for remote clouds no shift at all is seen. This subdivision of clouds shows clearly: The shift of the frequency distribution represents the radius effect, which generally depends on the degree of pollution. The radius effect is enhanced in more highly polluted regions.

[49] Another characteristic of the absorption effect is visible in the range at about +30% in Figure 4c, which represents optically thick clouds. At +30% all curves are converging, indicating another shift in the other direction for the positive branch of the frequency distribution. This shift is also known from theory. The shift, which is stronger in polluted regions, is much less pronounced than the shift by the radius effect for the optically thin clouds is. The change, now for the optically thick clouds, again clearly depends on the degree of the pollution. This second shift is mainly induced by the saturation effect of the cloud reflectance, which appears for clouds that are sufficiently optically thick.

The absorption effect, which works against the radius effect, is responsible for this behavior of the frequency distributions. If we consider the reflectance and other optical properties of the cloud layer to be functions of only optical thickness and single-scattering albedo, it becomes obvious that for a fixed single-scattering albedo the rate of increase of reflectance decreases with increasing optical thickness. Just this effect is seen in our results for the positive branch between +20% and +30% deviation from mean with a reflectance in the range of nearly 80–90% (see Figure 4c).

[50] It should be noted that the shifts in frequency distributions for JF9699 and JF8184 show similar characteristics as for JF8589.

[51] In the case of very highly reflecting optically thick clouds there is negligible anthropogenic influence. This can be explained by optically thick clouds possibly in combination with an additional cloud layer above, i.e., cumulonimbus combined with a thick cirrus layer.

[52] For very low reflectance in source regions of pollution, i.e., for strongly polluted areas with mean reflectances below 58% during winter, the frequency distributions include a secondary peak (see Figure 4c). The distributions show this peak at −40%, with a cloud reflectance that is below 25%. These remarkably low reflectance values most likely indicate clouds, i.e., subpixel cloudiness, within thick haze layers containing extremely high amounts of BC. The circumstance that the secondary peak disappears for the late 1990s supports the fact that brown haze occurred during the 1980s in Europe. Therefore we tentatively name this anthropogenic influence in the frequency distribution the black carbon peak.

[53] The black carbon peak is most pronounced during 1981–1984 for ND and JF, but it is also pronounced for JF8589. In the frequency distributions for ND8589, ND9699, and JF9699 such a pronounced secondary peak induced by haze does not become visible. The occurrence of haze confirms the high degree of pollution during the 1980s.

[54] Besides the secondary peak, called the black carbon peak, another point now becomes clear: Near the center the frequency distributions are generally more narrow in polluted regions. This is the result of a stronger radius effect for optically thinner clouds and a stronger absorption effect for optically thicker clouds working in opposite directions.

[55] The frequency distributions for summer are much narrower than those for winter. The mean standard deviation for the four summer months (MJJA) ranges from 11.3 to 12.5% (see Table 2). As in winter the values for Halle-Leipzig-Cottbus and Crakow-Katowice are increased as compared to the mean of the total area. However, during summer, in contrast to winter, variability follows the amount of SO_2 emission. The variability increases with increasing reflectance, which means that the SO_2 emission has a major influence on both the mean reflectance and the variability.

4. Discussion and Conclusions

[56] Our results show the radius effect (cloud albedo increase with pollution increase) over Central Europe in a region with a high density of power plants and traffic. This conclusion is derived through analysis of cloud reflectance frequency distributions from long-term satellite measurements.

[57] Unfortunately, a comparison of our results with the those of the International Satellite Cloud Climatology Project (ISCCP) is not advisable because of, first, the quite different spatial resolutions of the data sets. Second, a direct comparison with the ISCCP statistics http://isccp.giss.nasa.gov) is hampered by the distinct procedures for cloud classification.

[58] However, earlier studies dealing with satellite data and with in situ measurements confirm the strong influence of the elevated point sources on cloud albedo. There is, first, the work by *Rosenfeld* [2000], who derived smaller droplets within the dispersion plume of point sources from individual satellite scenes. The radius effect was detected for power plants as well as for lead smelters and oil refineries. Further, there are airborne in situ measurements made downwind of a coal-fired power plant near to Dresden in eastern Germany [*Keil et al.*, 2002]. The analysis of aerosol, microphysical, and solar radiative parameters within the dispersion plume generally confirms an albedo increase. However, despite enhanced aerosol particle and cloud droplet number concentrations, it was not possible to infer a decrease in effective cloud droplet radius by in situ measurements, which could be caused by additional water vapor emitted by the elevated stacks.

[59] In our study, a very large amount of data yielded a clearly modified shape of the frequency distribution in a strongly polluted region, which is well explained by the radius effect for optically thinner clouds. This long-term change during the past decades induced by pollution can be reconstructed in more detail as discussed below.

[60] Generally, the indirect aerosol effects have a strong influence on the variability of cloud reflectance. The variability increases when both less polluted and strongly polluted clouds are contributing. The higher variability can be achieved by a high BC content lowering the reflectance of strongly polluted clouds. However, effective secondary aerosol formation, leading to increased reflectance in the absence of a dominating absorbing component, can also be the reason. Besides, our data show that the influence of air pollution on cloud reflectance mainly depends on height and stability of the boundary layer. Additionally, the height of the cloud base and cloud top may have an influence. Explanation of our results can be made by considering the factors above.

[61] During ND the European boundary layer is more unstable and humid. Thus the impact of pollution on cloud formation and reflectance is more effective. Enhanced mixing, in particular, establishes the efficient vertical transport of particles into the cloud level. It can be assumed that as compared to JF, clouds, e.g., cumulus clouds, in ND have higher vertical extension, have a higher liquid water path, are optically thicker, and have higher vertical velocities. For cumulus clouds the inflow of pollutants is strong at the cloud base. However, the wet deposition processes, i.e., in-cloud and below-cloud scavenging, are effective in removing aerosols. Therefore, for cumulus clouds, a weaker indirect aerosol effect as compared to stratus clouds should be expected under the conditions of a higher PM load in the boundary layer.

[62] In order to make it visible we separated our data into stratus and cumulus cases by use of cloud observations in the strongly polluted area of Halle-Leipzig. Our hypothesis

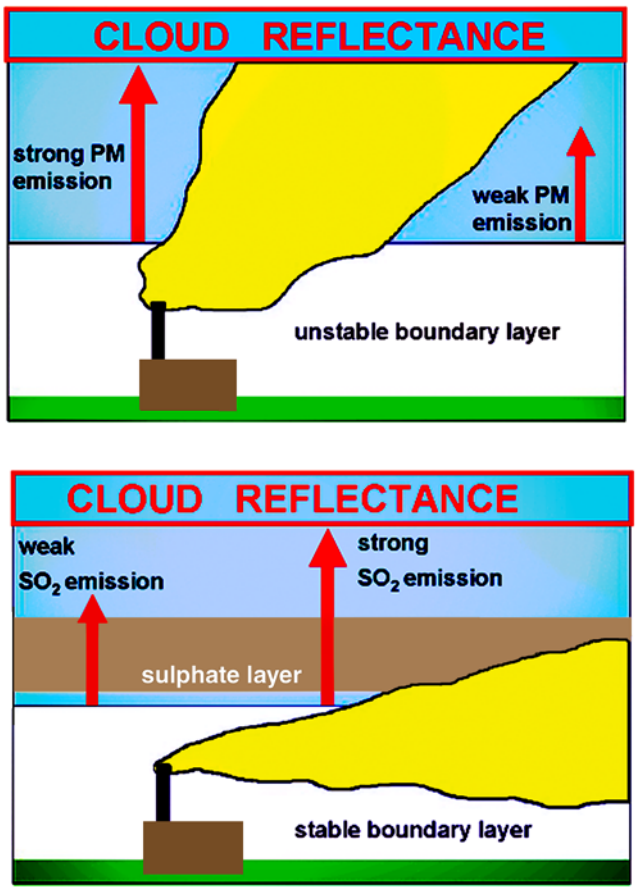

Figure 5. Schematic illustrating the different mechanisms of the so-called first indirect aerosol effect, i.e., the radius and the absorption effects, in unstable and stable boundary layers (plume of air pollutants originating from elevated sources in yellow, cloud layer in blue, and sulphate layer in brown). In the unstable boundary layer, aerosol particles originating from the surface can directly reach the cloud level by turbulence. The radius effect is working efficiently for strong primary particle emission. In stable boundary layers the dry deposition process prevents a strong effect by primary particles while the secondary aerosol formation becomes the major process. Under these conditions the formation of sulphate layers is pronounced. The development of haze layers is favored during weather situations with weak gradients, with low wind speed, and in absence of strong turbulence. The aerosol particle number can be enhanced by sulphate formation via the aqueous phase. An alternative process is the condensation of gaseous sulphuric acid on microparticles. Consequently, the cloud reflectance is increased by the radius effect during strong emissions of aerosol precursor gases.

is confirmed: The reflectance of stratus clouds for ND8184 is increased by more than 13%, while cumulus clouds show a much lower increase of 4% as compared to JF8184 (see Figure 5 and Table 3). This indicates the much more pronounced self-cleaning capacity of the atmosphere by cumulus clouds near the sources of pollution if wet deposition is active in unstable boundary layers.

[63] During the winters of the late 1980s the occurrence of enhanced indirect effects is shifted to JF. Now, in the case of more stable atmosphere, both cloud types stratus and cumulus are affected by haze layers. This is seen by comparing the data for cumulus and stratus clouds during the SO_2-rich emission episode of 1985–1989 (Table 3; also Figure 5) in much colder atmospheres. In contrast to ND the stable conditions are more frequent during the late winter months JF. This results in lower boundary layer heights and a clearly reduced vertical transport of particles. Despite the low mean cloud base, in many cases an increased dry deposition prevents the transmission of aerosol particles into the cloud layer. If cold cumulus clouds occur, the liquid water content is approximately several times lower than that for the more extended cumulus in the warmer months ND. Just this, referring to a low optical thickness, is the precondition for the enhanced radius effect. In addition, several shorter life-cycles of cold shallow cumuli might be rather effective in accelerating the aging of aerosols.

[64] Therefore our data show strong indirect aerosol effects during JF occurring for both layer clouds as well as cumulus clouds. Since the majority of cloudy atmospheres in late winter (JF) are accompanied by more stable stratification, a major influence of directly emitted larger particles can be excluded. Indeed, as Table 2 shows, a pronounced radius effect in JF is not seen for the episode 1981–1984. However, a maximum indirect aerosol effect takes place for the strongest precursor gas emissions from high stacks of power plants in the former GDR, pointing to the major influence of secondary aerosol formation.

[65] The coincidence with the high frequency of haze occurrence during JF8589 supports this explanation. In combination with a declining wet deposition in late winter, pollution also tends to accumulate within the boundary layer during the low mixing heights. Under these conditions, directly emitted fine BC particles from elevated stacks can contribute as the absorbing component. Furthermore, calm weather situations during late winter favor the aging of aerosols and lead, consequently, to more hydrophobic particles.

[66] The haze layers we detect are most likely sulphate dust layers, which occur in the absence of strong turbulence. Generally, the enhancement of aerosol particle number

Table 3. Mean Cloud Reflectance Derived From AVHRR Channel 2 Data for Days With Cloud Observations of Low Stratus or Cumulus Over the Area Around Halle and Leipzig (10.0°–15.3° Longitude and 50.8°–52.3°N Latitude) for the Winter Periods JF and ND From 1981 to 1984, 1985 to 1989 (Excluding Data From 1987), and 1996 to 1999[a]

	1981–1984	1985–1989	1996–1999
Stratus ND	70.9 (21.3)	57.8 (16.8)	58.2 (17.9)
Stratus JF	57.5 (20.3)	65.0 (14.9)	58.3 (11.1)
Cumulus ND	59.8 (19.9)	59.0 (18.2)	54.3 (17.9)
Cumulus JF	55.8 (20.5)	62.6 (17.9)	57.1 (16.0)

[a]Mean cloud reflectance is given in percent. The standard deviation is given in parentheses. ND, November and December; JF, January and February.

might be the result of sulphate formation via the aqueous phase. However, the condensation of gaseous sulphuric acid on microparticles should also be considered. In ND, however, a larger part of the pollutants is wet deposited, and fewer haze layers are resulting at the cloud level since the condensation nuclei are well mixed in the boundary layer. The haze layers above well explain the satellite measurements, which show a pronounced radius effect dominating the absorption effect, as seen by the highest reflectance and the moderate variability for episode JF8589.

[67] We conclude that strong pollution as seen for JF8589 generally tends to narrow the frequency distribution of cloud reflectance. However, the stability of the atmosphere determines the magnitude of particle transfer into the cloud level, and therefore the variability of cloud reflectance is increased because of the variability of the turbulence. The latter effect leads to a slightly higher standard deviation during the early 1980s as compared to the late 1980s. On the other hand, a higher difference between the mean JF and mean ND reflectance explains a higher JFND variability. As discussed above, a higher difference in cloud reflectance (Table 2) is mainly due to the increase of pollution during ND8184 and JF8589. While the variability increase for ND8184 is due to increased PM emissions, higher SO_2 emissions are the main cause during JF8589. If we compare this with the lower mean reflectance induced by higher BC content in aerosol during the early 1980s, then the much higher coefficient of variation for JFND8184 (Figure 3a) is well explained.

[68] During summer, the SO_2 emissions and thus the secondary aerosols largely determine the indirect aerosol effect, with dominance of sulphate particles. Experiments confirm that the contribution of BC to total particulate matter in remote areas, i.e., the Baltic Sea, is found to be nearly 2 times lower than it is in winter [*Kusmierczyk-Michulec et al.*, 2001]. The secondary aerosol formation can take place in the higher part of the boundary layer, while the higher PM concentrations occur much nearer to the surface. The consequence is that BC is much less efficiently incorporated into clouds. Because of the low BC content the variability of cloud reflectance is only slightly increased because of a limited absorption effect.

[69] We conclude that our study reveals a clear indication of impacts of air pollution on cloud reflectance over parts of Europe during the recent two decades. In summary, our results show the following: (1) The emissions of PM, including BC, and aerosol precursor gases have a pronounced influence on the cloud reflectance and its variability. (2) The cloud reflectance and its variability are determined by the radius effect, the absorption effect, and the saturation effect due to absorbing aerosols. (3) The so-called first indirect aerosol effect, i.e., the effect of a smaller droplet radius and stronger absorption, can be established via different pathways: under more unstable conditions by primary particle emission and in stable boundary layers through secondary aerosol formation. (4) The BC content is the major factor influencing the variability as seen by strong shifts of the frequency distributions to lower reflectance in polluted regions during winter. (5) A high concentration of secondary aerosols leads to a strong radius effect and shifts the frequency distributions to higher reflectance during summer, as well as during winter under stable boundary layers. (6) Haze layers occurring in winter confirm the high degree of pollution during the 1980s but have only a marginal influence on the overall variability.

[70] Besides this, our results still show a radius effect during the winters of the late 1990s. While haze is absent, the frequency distributions indicate a slight increase of variability from ND8589 to ND9699. This could be due to increasing emissions of fine particles with a strong absorbing component.

[71] Since the idea of this study was to detect the characteristics of indirect aerosol effects for widely suppressed interannual variability, further studies, including an evaluation of higher-resolution ISCCP data, are needed in order to estimate the climate forcing induced by anthropogenic aerosols.

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