Observational evidence for human impact on aerosol cloud-mediated processes in the Baltic region

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Olaf Krüger[1,2]

[1] Tartu Observatory,
61602 Tõravere, Tartumaa, Estonia

[2] Institute of Physics,
University of Tartu,
Ülikooli 18, Tartu 50090, Estonia;

e-mail: olaf.krueger@to.ee

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Abstract

Knowledge about aerosol cloud-mediated processes is important for judging climate change in Europe during recent decades. Here, some observational evidence for anthropogenic influences is described and discussed. The emphasis is laid on the effects of the large emissions of sulphur dioxide and particulate matter during the 1980s in Europe and the subsequent strong decrease in the 1990s. In addition, an analysis of the dependence of aerosol cloud-mediated processes on atmospheric circulation patterns (Großwetterlagen) is presented.

1. Introduction

Aerosol properties as well as cloud albedo are very uncertain forcing agents (IPCC 2007). However, while the planet's additional greenhouse effect is increasing, there are only a few observations indicating the impact of anthropogenic aerosols on clouds, e.g. Ackerman et al. (2000), Ramanathan et al. (2001), Krüger & Graßl (2002, 2004). This could be due mainly to the heterogeneity of source strengths, the short residence time and the multitude of chemical and physical processes that characterise aerosols. The greatest

uncertainty arises from the impact of variable aerosol particle numbers and the aerosol composition on cloud cover and the optical properties of clouds. Theoretical investigations underscore the fact that the influence of aerosol particles on radiative fluxes in cloudless atmospheres is negligible neither in the solar nor in the terrestrial spectral region. Within clouds aerosol particles may make a substantial contribution to heating rates in the solar part of the spectrum, while cloud albedo is a function of aerosol particle numbers and their chemical characteristics (Graßl 1978). Consequently, the cloud radiation field modified by aerosol changes is an important, wide-open issue, which needs to be addressed in estimates of global and regional radiation budgets.

Unlike the well-known positive radiative forcing caused by increased concentrations of long-lived greenhouse gases, anthropogenic aerosols can have different consequences for the radiation budget. They can either warm or cool the earth/atmosphere system. Hence, the sign of direct aerosol forcing for cloudless atmospheres is determined by both backscattering and absorption, which may vary considerably in the vertical. The reflectance of the underlying surface also plays an important role. If the surface is non-Lambertian, the bidirectional reflectance distribution function (BRDF) has to be considered (Kriebel 1978). The apparent reflectance, i.e. the reflectance of a natural surface modified by Rayleigh scattering and the overlying aerosol layer(s), varies with optical thickness and type of aerosol. The wavelength-dependent influence of aerosols ranges from an increase for low reflectance to a decrease in the case of a strongly absorbing component. Greater absorption is characteristic of urban aerosols, which usually contain much more black carbon (BC) than continental aerosols. A lowering of reflectance, resulting in a warming effect at the surface, can take place for a strongly absorbing component in the aerosol above a highly reflecting surface like white sand, snow or ice (Krüger & Fischer 1994).

Once deposited on the surface, absorbing aerosols can also alter surface reflectance. Analysis of BC in snow water shows mean values of 30 ppb (parts per billion by mass; equivalent to ng/g or μg per litre meltwater) in fresh, non-fresh, firn and windblown snow, even in the Arctic, indicating its relevance to global warming (Noone & Clarke 1988). Values at rural sites, e.g. in Lithuania, often exceed 100 ppb with peak values of 150 ppb during the cold season (Armalis 1999).

The Fourth Assessment Report (AR4) of the IPCC indicated that the mean global radiative forcing caused by the direct aerosol effect amounts to about -0.5 W m^2. The cloud albedo effect, which is least well understood by scientists, is estimated to be negative, reaching about -0.7 W m^{-2} in the global mean (IPCC 2007).

However, major uncertainties seem to be related to knowledge about carbonaceous aerosols. Bond et al. (2013) stated that the global atmospheric absorption attributable to BC is too low in many models and should be increased by a factor of almost 3. Those authors found the best estimate of industrial-era climate forcing of BC including all forcing mechanisms to be $+1.1$ W m^{-2}. However, they concluded that uncertainties in net climate forcing from BC-rich sources are substantial, which points to aerosol cloud-mediated processes for BC and co-emitted organic carbon.

Observations confirm that at different scales characteristic atmospheric perturbations become dominant, depending on solar irradiance and on their location in the earth-atmosphere system. For the Baltic Region the following processes are expected to be relevant:

- At the cloud scale, the interaction of cloud and aerosol processes determines the initial concentration and size of droplets. In many regions aerosol-cloud interactions are perturbed by increasing amounts of anthropogenic aerosol particles. In this respect, changes in cloud albedo, cloud lifetime and the amount of precipitation exert the greatest influence.
- At the regional scale, pronounced indirect aerosol effects are expected to occur in areas with strong anthropogenic release of particles, i.e. the so-called polluted regions. This may modify the additional greenhouse effect considerably. The influence of aerosols on clouds could dominate other perturbations, dampening or amplifying cloud-radiation feedback, which considerably reduces the relative importance of any other forcing agent at this scale. In the case of a more heterogeneous aerosol perturbation, the regional radiation regime in an initial environment will react with a more variable radiation budget, in contrast to the much more homogeneous warming by long-lived greenhouse gases. This heterogeneity of aerosol effects is also due to the pronounced dry and wet deposition processes in the atmospheric boundary layer. The general influence of aerosols may be identified in data records for albedo, solar irradiance, temperature, precipitation and cloudiness.

For Europe knowledge about the emissions and concentrations of air pollutants, and in particular information on aerosols and their precursor gases, is quite comprehensive. In addition, measurements and model calculations indicate the strong variability of this pollution plume owing to changing emissions, chemical transformations, deposition and long-range transport of manifold species, all depending on season and weather type (see e.g. Eliassen & Saltbones 1983, Krüger & Tuovinen 1997, EMEP 2004, Schaap et al. 2004, van Dingenen et al. 2004, Putaud et al. 2004).

2. Surface solar radiation

During the late 1980s, enormous amounts of particulate matter and aerosol precursor gas emissions, such as sulphur dioxide, nitrogen oxides and ammonia, made a strong contribution to the aerosol load over Europe. The extraordinarily high sulphur dioxide emissions in the former German Democratic Republic (GDR), which amounted to even more than 5 Tg per year, were of major importance to secondary aerosol particle formation. The sizeable contributions from elevated point sources around Halle, Leipzig and Cottbus resulted in pronounced spatial differences of sulphur dioxide (SO_2) and particulate matter (PM) concentrations in air. Such an increase in air pollution lead to reduced extinction and altered cloud optics.

In Germany Liepert & Kukla (1997) found a statistically significant decrease in the mean annual surface global solar radiation between 1964 and 1990 under completely overcast skies. This result can be potentially explained by an increase in cloud optical thickness, changing cloud types, or by human impact on aerosol cloud-mediated processes.

The collapse of the Eastern Bloc in 1989 led to significant reductions in industrial activities and thus atmospheric pollution. A pronounced declining trend was observed in the so-called 'Black Triangle'; this name refers to the enormous damage to human health and ecosystems caused by soot. This area, covering the southern part of Saxony (Germany), northern Bohemia (Czech Republic) and south-western Lower Silesia (Poland), is a prominent example of the extensive use of lignite deposits in Europe.

Stjern et al. (2011) analysed the visibility changes in the 'Black Triangle' between 1983 and 2008. They confirmed that the strong reductions in SO_2 and PM emissions in central Europe, i.e. a 90% decrease of SO_2 emissions and a 72% decrease of measured sulphate concentrations, improved the mean horizontal visibility in the 'Black Triangle' from 11 to 27 km between 1983 and 2008.

3. Cloud albedo

Satellite data indicate clear changes in cloud albedo for the strong pollution episode in Europe. A cloud albedo effect can be attributed to changing emissions of sulphur dioxide and particulate matter. This effect is based on an analysis of a reprocessed set of satellite measurements from 1985 to 1999 (Krüger & Graßl 2002). Two episodes of cloud reflectance, in the late 1980s and the late 1990s, over the central European main emission area have been compared.

The major result of the study was a pronounced cloud albedo decrease of about 2% from the late 1980s to the late 1990s owing to the decrease in

aerosol precursor gases. During winter in source regions of anthropogenic PM emissions, the cloud reflectance is smaller by more than 5%, which in addition points to an absorption effect caused by black carbon in clouds. Comparisons with emission data as well as model results of long range transport over Europe support the conclusion that aerosol cloud-mediated processes are responsible for significantly changed cloud optical properties.

The radiative forcing based on these data for the classical Twomey effect (Twomey 1974) amounts to about 1.5 W m^{-2} from the late 1980s to the late 1990s. Furthermore, during winter a radiative forcing of about 3 W m^{-2} due to the absorption effect, i.e. the albedo reduction of clouds (Graßl 1975), was estimated for both the late 1980s and the late 1990s.

Further insights into cloud albedo changes can be obtained by considering different European atmospheric circulation patterns (Großwetterlagen). Therefore, the satellite data are evaluated separately for different circulation conditions. A promising way is to consider Großwetterlagen for analysis. Here, we use the catalogue by Gerstengarbe & Werner (2005) containing the daily European atmospheric circulation patterns, provided by the Potsdam Institute for Climate Impact Research, together with the German Weather Service.

The atmospheric circulation patterns, which are defined as the mean air pressure distribution over an area at least as large as Europe, are eminently suitable for further subdividing the satellite data over central Europe. The original classification scheme considers three circulation groups comprising 10 major types and 29 sub-types plus undetermined cases. Here, the two major groups, zonal and meridional circulation, are taken into account to assess the influence of aerosols on cloud albedo.

The zonal circulation group definition in the catalogue is: 'High sea level pressure covers subtropical and lower middle latitudes and low sea level pressure exists in the sub-arctic and higher middle latitudes. The upper airflow is west to east. Cyclone tracks run from the eastern North Atlantic into the European continent. The zonal circulations include all circulation types 'West'.'

When using the data set by Krüger & Graßl (2002) the zonal circulation group during winter exists for 40% of the data for JFND8589 and 30% for JFND9699; but only 25% for MJJA8589 and 27% for MJJA9699.

The definition of the meridional circulation group by Gerstengarbe & Werner (2005) is: 'Stationary blocking high-pressure centres at sea level are characteristic of the meridional circulation. Depending on the location of the sea level pressure centres and the resulting main flow directions to central Europe, the types 'North', 'South' and 'East' can be distinguished.

In addition, all troughs with a north to south axis are classified as meridional circulations. The major types 'North-East' and 'South-East' are also included in the meridional circulation group because they normally coincide with blocking highs over Northern and Eastern Europe.'

The meridional circulation group during winter is due to 25% of the satellite data (Krüger & Graßl 2002) for JFND8589 and 35% for JFND9699, and during summer 38% for MJJA8589 and 39% for MJJA9699.

The analysis confirms the same tendencies of cloud albedo changes independently of the circulation group. The changes are in line with the results presented in Krüger & Graßl (2002) and Krüger et al. (2004). The cloud albedo for the zonal and meridional circulation groups during winter (JF and ND) is shown in Figure 1. The tendencies for the zonal as well as the meridional circulation groups appear to be conspicuously connected to PM emission changes on the one hand (during ND) and SO_2 emission changes on the other (during JF).

Firstly, a decrease in reflectance from the early 1980s to the late 1990s occurs during early winter (ND). The albedo decreases primarily following the reduction in PM emissions in Germany. It is more pronounced for the meridional circulation.

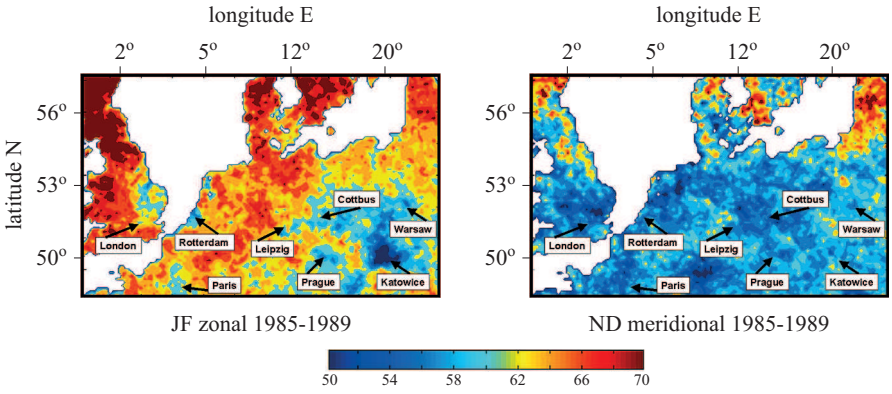

Figure 1. Mean cloud reflectance of low and medium level clouds derived from AVHRR channel 2 data over parts of Europe, given in percent. Data are shown from longitude 5.0°W to 24.0°E and latitude 48.5° to 57.5°N for zonal circulation during the winter period JF (January, February) and meridional circulation during ND (November, December) from 1985 to 1989 (excluding data from 1987). The grid size is 0.250 degrees longitude and 0.1250 degrees latitude. The area covers parts of the UK, Denmark, Sweden, France, Benelux, Germany, Poland, the Czech Republic, the Baltic countries and Ukraine. The North Sea and the Baltic Sea are shown in white. Strongly polluted areas are indicated. The catalogue of daily European atmospheric circulation patterns (Großwetterlagen) by Gerstengarbe & Werner (2005) was used for this analysis

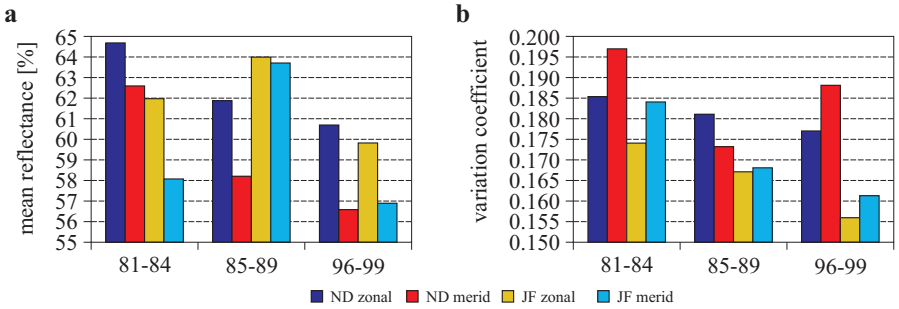

Figure 2. a) Mean cloud reflectance of low and medium level clouds for three time periods derived from AVHRR channel 2 data over parts of Europe, given in % (left-hand diagram). The values include the data from longitude 5.0°W to 24.0°E and latitude 48.5° to 57.5°N for the two-month periods ND (November, December) and JF (January, February). A subdivision is made for zonal and meridional weather types. b) The diagram to the right shows the corresponding coefficients of variation

The highest cloud albedo in ND during the early 1980s pollution episode can be explained by the existence of the radius effect (Twomey 1974). Enhanced turbulence during ND, as compared to JF, may well have favoured the effective lifting of primary aerosols to cloud level. The cloud albedo during ND8184 as compared to ND9699 was 4% higher for zonal circulation and even 6% higher for meridional circulation (see Figure 2).

Secondly, the magnitude of the cloud albedo in JF for both circulation groups tends to follow the level of SO_2 emissions, which originated mainly from large power plants in the former GDR (as described by Krüger et al. 2004). The highest value of the albedo is for JF8589, which points to the major influence of secondary aerosols. As before, the changes for the meridional circulation group are stronger. The most likely explanation is that the episodes in late winter with the more often stably stratified atmospheres favour the formation of sulphate layers, i.e. haze, which in turn enhance the cloud albedo through the radius effect (Krüger et al. 2004).

Thirdly, the trend in cloud reflectance variability seems to be highly influenced by the PM emissions, because of the higher BC content, which can lead to greater absorption and a lower cloud albedo. In addition, secondary particles are contributing to the overall variability through an albedo increase (Figure 2).

Another interesting result becomes apparent when we compare the change in variability for both circulation groups. Only for zonal circulation does the variability in ND and JF jointly decrease: for meridional circulation the variability of cloud reflectance decreases from JF8589 to JF9699 but increases from ND8589 to ND9699. This could be due, for example, to

two processes: an increase in BC emissions within the study area, and the advection of pollution from southern or eastern Europe outside the study area under consideration.

The major tendencies described above for three four-year episodes for the zonal and meridional circulation classes are well reproduced even if we analyse only two-year episodes. The result points to the dominant influence of pollution and not to changes in circulation.

In winter the most pronounced radius effect occurred during JF for both the zonal and meridional circulations. This can be explained by the influence of sulphate layers in the more frequently stable atmospheres. The maximum albedo decrease of 7.8% is due to the meridional circulation type in highly polluted regions, which show a comparably low JFND8589 reflectance. A more detailed analysis for the area around Leipzig reveals that the cloud albedo effect is stronger for stratus clouds than for cumulus clouds (Krüger et al. 2004).

The results for summer also support the conclusion of an anthropogenic influence over the most polluted part of central Europe including Germany, Poland and the Czech Republic. The highest decrease in reflectance, by more than 4%, occurred in areas with the highest SO_2 concentrations during the late 1980s. Remote regions show, as for winter, a much weaker decrease in reflectance of only 1%. The stronger changes for the meridional circulation could be due to a lesser air mass exchange and a subsequent accumulation of pollutants in the atmospheric boundary layer. A very interesting result emerges for coastal areas: the cloud albedo is increasing towards the 1990s but decreasing in areas of maximum sulphate concentration during the 1980s. This phenomenon is seen in the more frequent unstable weather situations during MJ, when advection of aerosol particles from source regions to the coastal areas is enabled by meridional circulation. The result may suggest that the number concentration of fine particles in parts of central Europe may have increased from the late 1980s to the late 1990s.

4. Cloud brightness temperature

The identification of cloud albedo changes as a consequence of pollution changes over Europe provided the motivation for investigating whether anthropogenic aerosol particles could even change cloud dynamics. The general hypothesis was that if anthropogenic aerosols do exert an influence on cloud dynamics, this should be detectable in the areas of strongest cloud albedo changes. Since a change in cloud dynamics will always be accompanied by a change in cloud brightness temperature, the scientific

question was whether the brightness temperature of cloud tops changed in the strongly polluted regions of Europe.

Indeed, Devasthale et al. (2004) detected a decrease in brightness temperature for stronger air pollution in central Europe during the late 1980s. The cloud brightness temperature changed in low- and medium-level and convective clouds. During episodes of strong anthropogenic emissions in Europe, the cloud-tops over and around polluted regions are higher, and their temperatures exhibited greater variability. In the area shown in Figure 1 the cloud top temperature increased during summer by 4.4K over the land and 1.6K over the sea. During winter the increases over the land were somewhat smaller (by 3.7K). During the summers of the late 1980s, the brightness temperatures of low- and medium-level clouds close to emission sources changed by 2.9K and those of convective clouds by as much as 5.2K. This signifies the evident human impact of aerosol cloud-mediated processes in the thermal spectral range.

The impact of ship emissions on cloud properties over coastal areas was also investigated using the same data set (Devasthale et al. 2006). Whereas land-based emissions were decreasing in central Europe, emissions from ships were increasing. The pollution from shipping routes in the English Channel and from the top three polluting harbours in Europe caused an increase in cloud albedo and a corresponding decrease in cloud top temperature; both parameters were more variable over coastal areas.

The debate is continuing as to whether the cloud property changes induced by ship exhaust emissions (commonly referred to as 'ship tracks'), first observed by Conover (1966), are due to a decrease in droplet size or to an increase in the cloud liquid water path through additional droplets. Radke et al. (1989) pointed out that the latter process could well explain this finding, because the number of condensation nuclei is generally limited over the ocean, which is not the case over the land. Since large numbers of Aitken nuclei can be formed in the exhaust, ocean-going vessels could easily contribute to the anomalous formation of Aitken nuclei. Conover (1966) specified the critical conditions for this to happen. In particular, convectively unstable situations from the surface up to a stable, low-level layer, as well as a slight supersaturation at the top of the convective layer, presumably deficient in cloud nuclei, favour the observed anomalous cloud lines. These ship tracks have been widely used together with Twomey's theoretical work (e.g. Twomey 1977) to manifest the great importance of indirect aerosol effects in the climate system. Field experiments in marine stratocumulus clouds supported the above conclusions regarding the occurrence of indirect aerosol effects (Coakley et al. 1987).

5. Precipitation

Later in 1989, Albrecht (1989), also influenced by the finding of Radke et al. (1989), formulated the basis for the so-called second indirect aerosol effect in his theoretical work. He postulated that a reduction in drizzle increases cloud lifetime over the ocean because this process regulates the liquid water content and energetics of shallow marine clouds. This in turn would contribute to a cooling of the Earth's surface and could have enormous consequences for climate. Consequently, special emphasis was given to investigations on whether drizzle is suppressed in ship tracks. In 2000 this second indirect effect, often called the cloud lifetime effect, was detected by both a field experiment and satellite measurements. Ferek et al. (2000) were able to show by radiometric measurements and radar observations that increased droplet concentrations in ship tracks, accompanied by smaller droplet sizes, significantly alter the liquid water path. In observations of the Tropical Rainfall Measuring Mission (TRMM) over South Australia, Rosenfeld (2000) found the same result on the cloud scale: drop growth by collision is very effectively suppressed by anthropogenic aerosol particles originating from power plants, lead smelters and oil refineries. The same effect of cloud droplet size reduction together with a delay in the onset of precipitation was found over the Amazon during the Large-scale Biosphere-Atmosphere Experiment in Amazonia subproject on Smoke, Aerosols, Clouds, Rainfall and Climate (LBA-SMOCC), where it was shown in detail what an enormous influence thick smoke from fires can have on cloud microphysics (Andreae et al. 2004). Other comprehensive field experiments which contributed considerably to knowledge about aerosol cloud interactions are the Smoke, Clouds, Radiation-Brazil (SCAR-B) experiment (Kaufmann et al. 1998), the Tropospheric Aerosol Radiative Forcing Experiment (TARFOX), the Indian Ocean Experiment (INDOEX), the Aerosol Characterization Experiments (ACE-1) (Bates et al. 1998) and ACE-2 (Raes et al. 2000) and the Aerosol Characterization Experiment in Asia (ACE-Asia). The planning of these field campaigns was stimulated by the presence of global fields of aerosol optical thickness derived from satellites (e.g. Husar et al. 1997) as well as by global model results (e.g. Langner & Rodhe 1991), which highlighted certain regions with conspicuously enhanced aerosol concentrations.

One of these regions is the Indian Ocean. Here, INDOEX discussed another aspect of indirect aerosol effects: highly absorbing aerosol particles and their long-range transport over the ocean. Trade wind cumuli were moving within deep layers of dark haze. Based on these observations Ackermann et al. (2000) suggested that the reduction of tropical cloudiness by soot could represent another major effect of aerosols on clouds. Model

calculations showed that the typical decrease in relative humidity during the daytime driven by solar heating with a maximum around noon is enhanced by the presence of absorbing haze in the boundary layer. As compared to the influence of the 'white' non-absorbing aerosol, the absorbing haze leads to a higher temperature, a lower relative humidity and a shorter anvil lifetime. The process, which shows a clear diurnal cycle, is called the semi-direct effect or cloud burning.

Despite recent advances in aerosol-cloud-precipitation interactions (see e.g. Lohmann & Feichter 2005, Wood et al. 2011, Stubenrauch et al. 2013) there still exist major gaps in our knowledge about the processes involved (Stevens & Feingold 2009).

For Europe there are no indications for diurnal cloudiness cycles owing to the influence of air pollution. Nonetheless, there does exist a statistical analysis for the years 1991–2005, after the strong emission episode in the Black Triangle, which illustrates significant weekly periodicities in many variables such as temperature, daily temperature range, sunshine duration, cloud amount, precipitation, and precipitation frequency (Bäumer & Vogel 2007). Derived from both metropolitan areas and more remote stations, e.g. on the Zugspitze (altitude 2960 m), these findings may point to atmospheric dynamics on a larger scale rather than just directly to daily changes in the aerosol system.

Besides the weekly periodicities mentioned above, there are indications that the strong emissions of SO_2 and particulate matter during the 1980s in Europe affected precipitation processes. Stjern et al. (2011) found that pollution reductions in the Black Triangle caused a substantial increase in horizontal visibility of 15 km from 1983 to 2008. The results are based on an analysis of synoptic weather observations (SYNOP) from the European Centre for Medium-Range Weather Forecasts' (ECMWF) Meteorological Archive and Retrieval System. In addition Stjern et al. (2011) used gridded precipitation data sets from the Climate Research Unit (CRU) of the University of East Anglia and from the Global Precipitation Climatology Project (GPCP) and sulphate measurements from the European Monitoring and Evaluation Program (EMEP).

In contrast to the evident change in visibility, the authors found no sign of any influence of aerosols on total precipitation trends in Europe. However, the annual frequency of light precipitation events, i.e. precipitation as events with less than 0.5 mm in 12 hours, increased significantly. For the area of the Black Triangle alone, significant changes in both total and light precipitation frequency were found and were attributed to air pollution.

It is interesting that the trends analysed by Stjern et al. (2011) were more distinctive in summer. This is in line with the stronger summertime

cloud albedo effect and the stronger brightness temperature change found by Krüger & Graßl (2002) and Devasthale et al. (2004) for Europe.

6. Discussion

There is clear evidence of human impact on aerosol cloud-mediated processes during the 1980s. This influence is seen for cloud albedo, cloud brightness temperature and precipitation. The finding of an effect of aerosols on clouds in the thermal spectral range in addition to the visible spectral range is of major importance, because it further complicates the estimation of radiative forcing due to aerosol cloud-mediated processes. To date, however, there is no plausible explanation for the process chain involved in the anthropogenic change of cloud microphysics caused by aerosols.

Further, it can be concluded that the detection of changes in essential variables, i.e. cloud albedo, cloud brightness temperature and amount of precipitation, is most pronounced in the source regions of air pollutants. This might indicate the regional character of the processes rather than an influence on a global scale. However, the hydrological cycle and the radiation balance in Europe were involved.

In addition to the knowledge described above, there are significant observations which need further attention in future studies. These results are important constraints for evaluating the nature of cloud-mediated processes and to further quantify the magnitude of human impact on climate in Europe.

The first result is related to time series of global irradiance in Estonia (Eerme et al. 2010). Measurement data show a conspicuous low relation of global irradiance for overcast and clear conditions (G/G_{clear}) during the summers of the late 1970s and the 1980s. Furthermore, the characteristics for normalised surface global solar radiation seem to be conspicuously connected to the results for Germany by Liepert & Kukla (1997), which are mentioned above. In principle, cloud properties could have changed as a consequence of anthropogenic aerosols. However, if this is indeed the case, the origin of the impact has to be identified. Also requiring further investigation is whether both time series could be explained in a similar way by aerosol cloud-mediated processes in Europe.

Another result which needs to be taken into account when assessing the influence of European aerosol system on clouds is the behaviour of organic vapour emissions in Europe. Paasonen et al. (2013) found that rising biogenic organic vapour emissions in response to warming will enhance condensation on particles and their growth to the size of cloud condensation

nuclei. The authors specify the strongest negative feedbacks at the most northern and remote sites.

Moreover, phytoplankton related emissions, such as dimethyl sulphide (DMS) and volatile organic gases via their transformation into aerosol particles need to be considered for the Baltic Region. The subsequent formation of cloud condensation nuclei (CCN) (O'Dowd & de Leeuw 2007) can decrease the shortwave radiation flux at the surface and increase the reflected shortwave radiation flux at the top of the atmosphere as well as decrease the amount of precipitation (Krüger & Graßl 2011). Thus, biologically produced gases dissolved in oceanic waters and their subsequent emission into the marine troposphere play an important role and are likely to be involved in regional feedback processes in the climate system.

Therefore, we need to investigate the influence of changing anthropogenic and natural emissions (e.g. sulphur, black carbon and biogenic volatile organic compounds (BVOCs)) on regional climate. Modelling activities constrained by observations need to be focused on aerosol cloud-mediated climate in the Baltic Region. This ideally should include:

- Treatment of biogenic and carbonaceous aerosols.
- The change in biogenic emissions in a changing climate, including dimethyl sulphide (DMS).
- Effects of spatial gradients in solar heating (dependent on circulation types).
- Effects of higher CCN numbers and absorption on cloud albedo and precipitation.

Summarising the results presented above, it can be concluded that there is clear observational evidence for an anthropogenic influence on aerosol cloud-mediated processes over Europe.

The effects show individual characteristics for different atmospheric circulation patterns. The next steps need to combine atmospheric modelling and different observations synthesised in more detail, including the latest achievements from field studies aiming to analyse the European aerosol system (Kulmala et al. 2011).

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