Aerosol connections between three distant continental stations

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ABSTRACT

The present study is based on hourly aerosol particle and ancillary data taken at three stations in Northeastern Germany 150–200 km apart in the years 2009 through 2015 in order to investigate systematic process related differences that might show up when connecting their data with forward and backward air mass trajectories. The analysis of changes in the atmospheric aerosol during air mass transport between the stations focused on two low-pollution-to-high-pollution pathways of similar distance around 200 km. Despite rather different initial size distributions the increases in total concentration of particle number, volume, and black carbon agreed within 15%. Systematic variations were found in the different concentration increases as a function of time of day at which an interstation air mass transport took place that could be explained with related source and transport processes.

With Radar-based precipitation data at the stations and along connecting trajectories sub-cloud particle scavenging was investigated. At each station sub-cloud particle scavenging coefficients were determined as a function of particle size between 10 and 700 nm diameter, and as a function of precipitation sums. Median particle scavenging coefficients taken over the central part of the size range of the study strongly increased between 0.2 and 1 mm of precipitation and then planed out towards higher precipitation values. With the particle scavenging coefficients determined at the individual stations and precipitation data along connecting trajectories the sub-cloud particle scavenging analysis was extended to wet scavenging along the transport paths yielding median sub-cloud scavenging ratios over the range 0.2–25 mm of precipitation sums with a non-linear shape similar to that of the median particle scavenging coefficients at the individual stations. Whereas in light precipitation more than 80% of the initial particles in the considered range survived 150–200 km of transport, more than 50% was scavenged by more than 15 mm of precipitation during aerosol transport.

1. Introduction

With the advent of automated mobility particle size spectrometers, (e.g., Birmili et al., 1999) stable long-term time series of high resolution sub-micrometer particle number size distributions became possible. First, few individual stations with such instrumentation, (Baltensperger et al., 1997; Birmili et al., 2001; Mäkelä et al., 2000), and later, networks of stations were established, (Asmi et al., 2011; Birmili et al., 2016; Dal Maso et al., 2007; Tunved et al., 2003).

Concurrent measurements at several stations of an aerosol network stimulated the study of spatial variation of the near-surface aerosol, beginning on local scales within cities. Buzorius et al. (1999) studied the variation of aerosol number concentration within Helsinki, Finland. Within Stockholm, Sweden, Johansson et al. (2007) determined spatial distributions of particle number and mass concentrations. Tuch et al. (2006) investigated the correlation of particle number size distributions

between two stations 1.5 km apart in Leipzig, Germany. With the focus on particle nucleation the comparative scale was widened to 50 km by Wehner et al. (2007). On that scale Lammel et al. (2005) investigated chemical aerosol changes in the urban plume of Berlin, Germany. Within the project Midwest Interstate Sulfur Transformation and Transport (MISTT) the urban aerosol plume of St. Louis, MIS was followed with trajectories to a rural site 104 km downwind (Ellestad, 1980). On a statistical long-term base aerosol data from a European network of stations were connected with 850 mb trajectories within the project Long Range Transport of Air Pollution (LRTAP) (Eliassen, 1978; Eliassen and Saltbones, 1983; Lemhaus et al., 1986). With state-of-the-art aerosol station data and modeling tools the aerosol over Europe was mapped more recently within the project European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI), (Kulmala et al., 2011). During the last decades the rapidly growing air pollution of China has stimulated aerosol transport and transformation

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studies in which aerosol data at individual stations were connected with satellite data (e.g., [Qin et al., 2016](#)) or mesoscale atmospheric models (e.g., [Gao et al., 2014](#); [Jiang et al., 2012](#); [Ye et al., 2016](#)).

The rapidly growing network of sophisticated aerosol stations in Northern European and Baltic countries became the base of numerous studies connecting the stations with air mass history, trajectories, and models. [Komppula et al. \(2006\)](#) combined particle number size distribution measurements and simulations of aerosol dynamics at stations in Finnish Lapland demonstrating the dominance of new particle formation over particle sinks in aerosol moving over 200 km. With one-year aerosol data at four northern stations [Tunved et al. \(2005\)](#) estimated the aerosol turnover time particle turnover times in air moving from south to north to 1–3 days depending on their size. Analyzing aerosol transport in the opposite direction [Tunved et al. \(2006b\)](#) found the number concentration of particles in the Aitken mode to increase while the mode peak diameter of the number size distribution did not change. [Tunved et al. \(2006a\)](#) determined secondary particle formation over Finnish forest areas by classifying air mass trajectories arriving at rural stations based on their transport time over the boreal forest area. They found that the newly formed particles gradually grew in size during transport in response to biogenic emissions on the way. [Väänänen et al. \(2013\)](#) combined aerosol dynamics and air mass trajectories in an analysis of aerosol data from three stations located within a distance of couple hundred km at the northern edge of the boreal forest zone. They determined particle growth rates along the transport paths and found that condensational growth can be important in aerosol dynamics even when new particle formation is not evident. [Kecorius et al. \(2016\)](#) investigated increases in aerosol particle number concentrations due to inter-station sources over distances of 490 and 530 km. [Vana et al. \(2004\)](#) speculated about even more than 1000 km-connections of particle nucleation events.

In 2008 the German Ultrafine Aerosol Network, (GUAN), was established to improve the understanding of aerosol processes over Germany ([Birmili et al., 2009, 2015](#); [Größ et al., 2018](#); [Nordmann et al., 2009](#)). For the present study we utilized long-term aerosol data taken with hourly time resolution at three GUAN stations in Northeastern Germany 150–200 km apart. As unique additional information radar-derived hourly precipitation rates from the German network of 16 C-band weather radars, ([Bartels et al., 2004](#)), were added to the aerosol data set at the stations and along hourly forward and backward air mass trajectories connecting the stations. The aim of the study was to understand systematic atmospheric process-related differences, which might occur when connecting the aerosol data with air mass trajectories under different meteorological conditions.

2. Database

2.1. Aerosol in-situ measurements

Particle number size distributions were determined in the sub-micrometer size range, using TROPOS-custom-made mobility particle size spectrometers (MPSS). The instruments at Waldhof and Neuglobsow had been designed for a diameter range from 10 to 800 nm, whereas in Melpitz a dual MPSS was installed, covering an extended size range from 5 to 800 nm. For the present study the common diameter range 10–800 nm was utilized. The data acquisition and the yearly calibrations of the instruments followed the recommendations in [Wiedensohler et al. \(2012\)](#) and [Wiedensohler et al. \(2017\)](#), respectively. The temporal resolution of the measurements in Waldhof and Neuglobsow was 5 min and in Melpitz 20 min. The data inversion was done following the method described by [Pfeifer et al. \(2014\)](#).

Mass concentrations of particulate equivalent black carbon, (BC), were measured at all stations, employing Multi-Angle Absorption Photometers (MAAP- Thermo-Scientific model 5012). The principle of the MAAP had been described in detail in [Petzold and Schönlinner \(2004\)](#). Aerosol particles were collected on a filter, and the light

absorption coefficient was determined by radiative transfer considerations, including effects of multiple scattering and absorption enhancement due to light reflection by measuring this parameter at two angles from the filter. The determination of particle light absorption coefficients was based on the transmitted and reflected light components, which were defined by directly measured values of the light transmission, direct and diffuse backscattering. The particle mass concentration of optically equivalent BC was internally calculated, using a constant Mass Absorption Cross-Section of $6.6 \text{ m}^2\text{g}^{-1}$, ([Petzold et al., 2013](#)). Temporal resolution of the MAAP was one minute. The performance and inter-comparability of MAAPs had been investigated in [Müller et al. \(2011\)](#) showing a general uncertainty of approximately 10%.

2.2. Ancillary gas data

At the station Melpitz, NO_x was determined with a chemiluminescence instrument (type Horiba APNA 370). For this method, the reaction of nitric oxide (NO) with ozone (O_3) to excited nitrogen dioxide was utilized. The photons emitted, when leaving the excited state, were counted with a photomultiplier, (NO measurement). To measure also NO_2 in the gas stream, the air was passed over a molybdenum catalyst. This procedure yielded NO and NO_x , and from the difference the concentration of NO_2 was determined.

At the UBA stations Waldhof and Neuglobsow, nitrogen oxides were determined with a nitric oxide analyzer from Thermo Scientific (type: 42i-TL). The measuring method was chemiluminescence with additional photolysis converter. The NO_x instruments were calibrated on a regular basis, using a certified calibration procedure. The primary temporal resolution of the NO_x measurements was one minute. Overall measurement uncertainty was considered to be between five and 10%.

2.3. Trajectories

Three-dimensional trajectories were calculated arriving and leaving every hour at a height of 500 m above ground level at the three GUAN stations. The trajectories were calculated forward and backward for up to five days using the HYSPLIT4 model ([Draxler and Rolph, 2003](#)) with meteorological data from the Global Data Assimilation System with one-degree resolution (GDAS1). The meteorological fields were downloaded from the server at Air Resources Laboratory (ARL), NOAA (<http://ready.arl.noaa.gov>), where more information about the GDAS dataset can be found. Aerosol connections between the stations were established by means of the hourly trajectories. Whenever a forward or back trajectory arriving or leaving one station passed another station within 10 h and a maximum distance of 50 km the data at the two stations were flagged as connected. On average, the closest distance between trajectory and passed-by station was about 30 km. The number cases, which were flagged as connected depended on the extent of the database at the stations and on the general atmospheric circulation over the region. By utilizing forward and back trajectories each transport direction between a station pair was covered twice. Thus, numbers of connected cases between 1000 and over 10000 during our study period were reached, (see [Table 1](#)).

In [Fig. 1](#) the four possible trajectory connections in between two stations (starting at/near A, and ending near/at B) were drawn schematically on the axis “Distance” and “Transport time” between the stations.

[Fig. 2](#) illustrates this approach with 3684 specific hourly forward trajectories from Melpitz to Waldhof in red, and 7501 forward trajectories from Waldhof to Melpitz in blue, ending closer than 50 km from the other station within 10 h during 2009–2015. Except for a few outliers that reached the other station on tortuous paths within the given time limit the connecting trajectories covered rather homogeneously a broad corridor between the two stations.

Potential meteorological differences between inter-station aerosol connections were explored with the information “Großwetterlagen”,

Table 1
Characteristics of the three GUAN stations ME = Melpitz, NG = Neuglobsow, WA = Waldhof, and of trajectory connections in between. Maximum allowed trajectory distance from any goal station was 50 km. Travel times in between stations up to 10 h were allowed. Cases with precipitation required ≥ 1 mm precipitation during ≥ 3 h of transport.

ID	Data coverage	Distance between stations (km)	Average travel time between stations (h)	Number of trajectory-connected data	Transport cases with precipitation
ME	2009-01-01 - 2015-12-31	ME - NG: 180	ME to WA: 7.7 ME to NG: 7.9	ME to WA: 2411 ME to NG: 1666	ME to WA: 94 ME to NG: 170
NG	2011-04-18 - 2015-12-31	NG - WA: 157	NG to WA: 5.4 NG to ME: 7.9	NG to WA: 1013 NG to ME: 545	NG to WA: 165 NG to ME: 150
WA	2009-01-01 - 2015-12-31	WA - ME: 205	WA to ME: 7.3 WA to NG: 5.4	WA to ME: 3338 WA to NG: 3211	WA to ME: 613 WA to NG: 888

Fig. 1. Schematic presentation of the trajectory connections between two stations, (A, and B), in terms of nominal distance and nominal aerosol transport time $\pm \Delta t$ along trajectories between the stations. Forward trajectories are drawn in red, backward trajectories in blue. A^- indicates the station A at the time $-\Delta t$ before arrival of a backward trajectory at station B, i.e., at time $t = 0$. B^- indicates the station B at the time $-\Delta t$ before arrival of a backward trajectory starting at station A. A^+ indicates station A at the time $+\Delta t$ after start of a forward trajectory from station B. B^+ indicates station B at the time $+\Delta t$ after start of a forward trajectory from station A. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

(GWL), concerning the 29 large scale weather type classifications after Hess and Brezowsky for Central Europe, (Gerstengarbe and Werner, 1993), provided by the German Weather service for each day (<http://www.dwd.de/DE/leistungen/grosswetterlage/grosswetterlage.html>). During the period of the present study, (and in general), the five most frequent GWL were, (in ranking order), WZ = Westerlies, mostly cyclonic, TRM = Trough over Central Europe, BM = High pressure bridge over Central Europe, SWz = Southwesterlies, mostly cyclonic, and Wa = Westerlies, mostly anticyclonic.

2.4. Radar data

Hourly precipitation estimates were obtained from the RW product of the German Weather Service Radolan composite (Bartels et al., 2004; Weigl and Winterrath, 2009). The product was based on radar-derived hourly precipitation rates from lowest elevation scans of the German network of 16 C-band weather radars, at which these scans had been performed every five minutes. The scans were merged and regridded onto a regular $1 \times 1 \text{ km}^2$ grid. Hourly radar-based rain rates were estimated from clutter-corrected and quality-assured 5-min radar-reflectivities using a categorized Z-R relationship and aggregation in time. Finally, a weighting technique was applied to adjust the radar-derived hourly precipitation rates to neighboring observations of the German rain gauge network. The detection limit of the RW product was set to 0.1 mm/h. The RW product had been used successfully previously, e.g.,

Fig. 2. 3684 hourly forward trajectories during 2009–2015 from Melpitz to Waldhof in red, and 7501 forward trajectories from Waldhof to Melpitz in blue, ending closer than 50 km from the other station within 10 h of travel time. The stations are marked with colored diamonds. Cities with populations of 100,000 or larger are shown as black circles on the map with the diameters of the circles being proportional to their population. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

in ensemble precipitation forecasts (Ben Bouallègue and Theis, 2014), and comparisons of different approaches to fit log-normal mixtures on radar-derived precipitation data (Kronenberg et al., 2014). In contrast to local rain gauge observations, the RW product had the advantage to capture the spatial structure and extent of precipitation systems, the effects of which on the atmospheric aerosol were part of the present study. Finally, the Radolan RW product was interpolated to the station locations and to trajectory positions taking the closest corresponding RW grid value.

3. Results and discussion

3.1. Inter-station aerosol comparison

The most basic way of considering aerosol changes after air mass transport between two stations concerns changes in total number, (ΔNTO), or total volume, (ΔVTO). Due to widely varying aerosol and transport conditions the probability distribution functions, (pdf), of ΔNTO along the six transport cases cover a wide range from decreases of more than 5000 cm^{-3} to increases of the same order of magnitude, (see Fig. 3).

A first comparison of these pdfs and dry average PNSDs at trajectory-connected stations, (see Fig. 4), yielded some insight into pollution changes and aerosol processes along the related pathways. We

Fig. 3. Probability density functions, (pdf, %), of changes in total number concentration, (ΔNTO , cm^{-3}), after air mass transport along the six different paths: WA $\rightarrow$ ME, Melpitz to Waldhof: ME $\rightarrow$ WA, Neuglobsow to Melpitz: NG $\rightarrow$ ME, Melpitz to Neuglobsow: ME $\rightarrow$ NG, Waldhof to Neuglobsow, WA $\rightarrow$ NG, and Neuglobsow to Waldhof: NG $\rightarrow$ WA.

distinguished three regimes of transformations along the connected paths:

- Regime 1. Low pollution to high pollution, (WA $\rightarrow$ ME, NG $\rightarrow$ ME),
- Regime 2. High pollution to low pollution, (ME $\rightarrow$ WA, ME $\rightarrow$ NG),
- Regime 3. Low pollution to low pollution, (WA $\rightarrow$ NG, NG $\rightarrow$ WA).

The low-pollution-to-high-pollution regime one was characterized by an increase in particle number concentrations over a broad range from about 30 nm to the upper size limit of the study. Below the lower limit of ≈ 30 nm both concentration decreases, (WA $\rightarrow$ ME), and

increases, (NG $\rightarrow$ ME), did occur. Air mass transport in opposite directions occurred in the high-pollution-to-low-pollution regime two, which was characterized by nearly constant PNSDs above about 100 nm and substantial losses of particles below that diameter. At a lower pollution level the transport WA $\rightarrow$ NG showed the latter loss/gain situation even clearer, (cf. bottom panels in Fig. 4), whereas transport in the opposite direction, (NG $\rightarrow$ WA) on average showed no significant changes in PNSDs over ≈ 160 km of transport distance. In following sections systematic aerosol changes in between the stations will be explored.

3.2. Inter-station aerosol processes

The complete description of aerosol transformations along an atmospheric trajectory on the time scale of ten hours requires the use of a complex chemical/physical aerosol dynamics model such as presented by Tunved et al. (2010). The formulation and/or application of such a complex model was beyond the scope of the present study. Instead, we focused our analyses on the discussion of cases and processes, which we could hope to be able to describe with simple transformation models, such as inter-station aerosol particle formation, and sub-cloud wet scavenging. There were several factors with which to choose from the six possible transport cases in between the three stations: One, logistics, i.e., the available number of connections in between a pair of stations, (cf. Table 1), two, the apparent changes in PNSD between the stations, (cf. Fig. 4), and three, the meteorological conditions during inter-station transport, (cf. Table 1). We excluded from detailed analyses all cases in regime two because they involved substantial small particle losses during dry conditions that would require complex aerosol dynamics for their description, i.e., ME $\rightarrow$ WA, and ME $\rightarrow$ NG. With the same argument we also excluded case WA $\rightarrow$ NG. Our interest in sub-

Fig. 4. Particle number size distributions averaged over all hours with connected trajectories without precipitation along the transport path. The depicted transport pathways are Waldhof, (green) to Melpitz, (blue): WA $\rightarrow$ ME, Melpitz to Waldhof: ME $\rightarrow$ WA, Neuglobsow, (red) to Melpitz: NG $\rightarrow$ ME, Melpitz to Neuglobsow: ME $\rightarrow$ NG, Waldhof to Neuglobsow, WA $\rightarrow$ NG, and Neuglobsow to Waldhof: NG $\rightarrow$ WA. The average difference of the respective two size distributions is drawn in black. Error bars indicate two standard deviations about the means. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

cloud aerosol scavenging corroborated our argument against focusing on regime two because it would have been difficult to distinguish sub-cloud scavenging from other strong loss processes in these cases. Additionally, meteorology argued against utilizing the cases in regime two for wet scavenging analyses because they predominantly occurred in flow situations with high-pressure systems over Eastern Europe or Scandinavia entailing small chances of precipitation. With the latter argument we also excluded case NG → WA.

Before we analyzed specific transport cases we addressed the question: How meaningful is a comparison of aerosol data at station B with corresponding data at station A taken before a calculated trajectory connection between the two stations? In other words, are the apparent aerosol particle changes due to processes along the transport path or are they due to common aerosol changes on scales comparable or larger than the distance between the stations? To explore this issue we compared integral particle properties at the individual stations after transport time Δt with the respective aerosol properties before Δt , (cf. Fig. 1 for the definition of Δt). For this test and for the subsequent growth results in section 3.3.1 we limited the analysis to transport cases with non-negative ΔVTO along the transport path, i.e., $VTO(B(t + \Delta t)) - VTO(A(t))$. The results are collected in Fig. 5.

In both investigated transport cases, (WA → ME, and NG → ME), average increases in total number, (ΔNTO), and total volume, (ΔVTO), were far higher when comparing station B after transport to station A before transport than corresponding changes at the individual stations, e.g., $A^{+\Delta t} - A$ or $B - B^{-\Delta t}$. Even for ΔBC this comparison yielded the highest average values after transport from station B to station A, albeit within two standard deviations of the average value. We note, however, that on average, except for ΔNTO at station WA temporal changes were positive at the individual stations during the transport events, indicating that the atmospheric conditions at the stations were conducive to aerosol increases. We took these comparisons of temporal and inter-station changes as strong argument for interpreting the differences between station B and station A after transport $A \rightarrow B$ as aerosol increases along the transport path and not as general aerosol changes on scales of distance between A and B or larger and proceed to quantifying the transport related increases in particulate matter in the following section.

3.2.1. Increase in aerosol particles in regime one (low pollution to high pollution)

For the study of aerosol particle increase from low to high pollution sites we had the connected data sets from two pairs of stations available: Waldhof to Melpitz, (WA → ME), and Neuglobsow to Melpitz, (NG → ME). Here particle increases are quantified in terms increases in ΔNTO , and ΔVTO , (see Table 2).

Prior to a detailed case analysis we addressed the two questions: When do events with apparently connected flows occur and at what times were the strongest aerosol increases recorded? For both transport paths the event counts per month show a broad maximum from March through August with a peak in June, (not shown). In terms of maxima in

ΔNTO , and ΔVTO the two paths differ. Whereas the path WA → ME exhibits a clear maximum in both parameters in April with a substantial secondary maximum in ΔNTO in September, ΔVTO on the path NG → ME peaks already in February with secondary peaks in June and November while ΔNTO has its main peak in June and a broad secondary maximum during winter, (November through February).

We explored the typical weather conditions of apparently connected flows paired with aerosol increases with the related GWL data. The “Grosswetterlagen” TRM and BM have high frequencies of occurrence on both paths with TRM standing for a trough over Western Europe directing polar air masses to the stations, and BM standing for a high-pressure bridge over Europe, over the eastern flank of which, again, polar air is directed to the stations. Highest occurrence for the path WA → ME was recorded with GWL system NWA with a strong frontal zone in the direction NW to SE. For the path NG → ME the most frequent GWL system was NA, i.e., strong northerly airflow caused by a blocking anticyclone over Western Europe.

When we excluded all cases $\Delta VTO < 0$, and with precipitation at any of the stations and in between 2312 connected hours remained for the pair WA → ME, and 403 h for the pair NG → ME. It is remarkable that both, ΔNTO , and ΔVTO along the two very different paths, (NG → ME and WA → ME), with very different starting PNSDs, (cf. Fig. 5), but with similar length lead to increases in particle number and volume that differed by no more than $\approx 15\%$.

Two logarithmic normal distributions were added to the average PNSD of the constrained data at station A to simulate the aerosol increase on the way from station A to station B. With Microsoft Excel's® nonlinear Generalized Reduced Gradients, (GRG), solver the parameters of these two normal distributions were optimized in order to yield minimum average relative deviations of 12–17% between the modified PNSD at station A and that at station B. The parameters of the derived lognormal distributions quantifying the average aerosol particle growth on the two low-to-high-pollution paths NG → ME, and WA → ME were collected in Table 2. The parameters of the two lognormal additions indicated that both Aitken, (below 80 nm), and accumulation mode, (≥ 100 nm), particles were added to the initial distributions. Average PNSDs at start and end of the events are shown in Fig. 6 together with the starting PNSDs to which the two optimum lognormal distributions were added.

The latter simulations of the aerosol at the end of the transport paths showed that on both paths not only aerosol increases were recorded. Below ≈ 40 nm particle diameter losses occurred during aerosol transport to Melpitz. These apparent losses may reflect particle growth through coagulation, condensation or liquid phase reactions. It is remarkable, though that, despite different pathways, initial and final aerosol, (cf. Fig. 6), comparable particulate number, volume and black carbon were gained during aerosol transport from Waldhof and Neuglobsow to Melpitz.

So far we considered aerosol particle changes averaged over months to years. Several factors can cause considerable diurnal variations in aerosol particle changes during transport between stations. Traffic and

Fig. 5. Average changes of total number, (ΔNTO , cm^{-3}), total volume, (ΔVTO , $\mu\text{m}^3\text{dm}^{-3}$), and Black Carbon, (ΔBC , ngm^{-3}), during conditions of dry transport with $\Delta VTO \geq 0$ at the stations WA, NG, and ME, compared to corresponding inter-station changes during transport NG → ME, and WA → ME. Error bars show two standard deviations of the mean values.

Table 2

Rows one and two: Parameters describing average aerosol growth on the low-to-high-pollution paths Neuglobsow to Melpitz, (NG → ME), and Waldhof to Melpitz, (WA → ME) by means of two lognormal distributions and by differences in particle number, (dNTO), volume, (dVTO), and black carbon, (dBC). SRR = sum of inter-station precipitation in mm. Cases = number of trajectory-connected cases with the constraint dVTO ≥ 0. Rel. fit error = average rel. deviation between the average size distribution at ME and the lognormal fit; N_o = Number concentration of a lognormal distribution at the geometric mean diameter D_g ; σ_g = geometric standard deviation of the lognormal distributions. Row three: Corresponding parameters for aerosol transport WA → ME with ≥ 1 mm of inter-station precipitation for at least three hours.

Path	SRR mm	Cases	Rel. fit error, %	N_o , cm ⁻³	D_g , nm	σ_g	dNTO cm ⁻³	dVTO μm ³ cm ⁻³	dBC, μgm ⁻³
NG → ME	0	403	12	701	65	1.3	790	2.9	.52
				593	141	1.7			
WA → ME	0	2312	17	605	65	1.5	450	2.7	.34
				285	197	1.6			
WA → ME	≥ 1	613	3	1120	31	2.6	-275	0.3	0.03
	≥ 3h			232	184	1.6			

energy-related emissions will change during a day as well as the photochemical production of particle precursors and their condensation, transport pathway, and ventilation of the planetary boundary layer. For the low-to-high-pollution station pair WA → ME with the largest number of cases we display median aerosol particle changes for non-precipitation conditions as a function of local time of day at the start of connecting air mass transport in Fig. 7.

Changes in particle concentration below 26 nm, (ΔN_{26}), exhibit the largest diurnal variation spanning a factor of 3.9 of the daily average, with their maximum value for transports starting around 06:00 and their minimum for transports starting around 18:00 local time. Second largest variations occurred in total volume, ΔVTO , covering a factor of 2.2 of its daily average, albeit with a phase shift of nearly 12 h as compared to ΔN_{26} . We interpreted the maximum of growth in particulate volume during nocturnal aerosol transport as being caused by condensation of particle precursors that had been produced during the day. We suspected that the rather small diurnal variation in the change of total number, i.e., a factor of 0.9 of the daily average, was both a consequence of new particle formation, as indicated by ΔN_{26} , and ventilation of the planetary boundary layer. The latter process would also affect the daily changes covering a factor of two of its daily average, of the combustion related black carbon, (ΔBC).

3.2.2. Decrease particles in regime one (low pollution to high pollution)

Wet scavenging of atmospheric particles is a subject of intense ongoing research. The analysis of long time series of PNSDs taken at aerosol observatories, combined with environmental data, can contribute to the understanding of particle scavenging, in particular, sub-cloud wet scavenging, (e.g., Laakso et al., 2003). Our analysis began with the evaluation of precipitation events at the three stations. With the results at the individual stations we proceeded to the discussion of

Fig. 7. Median changes in total number concentration, (ΔNTO , cm⁻³), number concentration below 26 nm particle diameter, (ΔN_{26} , cm⁻³), total volume, (ΔVTO , μm³dm⁻³), and black carbon, (ΔBC , ng³m⁻³), along the transport path WA → ME of 2312 events with $\Delta VTO \geq 0$ as a function of the time of day at which the event started.

the effects precipitation occurring on the transport paths in between the stations.

3.2.2.1. Scavenging at individual stations. In the present study we identified sub-cloud scavenging events by requiring an amount of precipitation > 0.2 mm per event with event lengths between three and 24 h. With these constraints we identified a total of 960 scavenging events in the time series at the three stations. The median length of the events was four hours, and median precipitation per event about 3 mm. The probability distribution function, (pdf), of the precipitation amount per event in Fig. 8 looked very similar at all three stations. Traditionally, scavenging coefficients Λ , (s⁻¹), are derived by assuming the scavenging to be proportional to both the length of the

Fig. 6. Average particle number-size distributions at start and end of transport WA → ME, and NG → ME during dry conditions with $\Delta VTO \geq 0$. Curves in black give respective starting size distributions with optimum added lognormal particle number size distributions according to Table 2, simulating aerosol growth during transport. Error bars show two standard deviations of the mean values.

Fig. 8. Probability distribution function, (pdf, %), of precipitation events as function of the precipitation per event, (mm), at the three GUAN stations Melpitz, (ME, blue), Neuglobsow, (NG, red), and Waldhof, (WA, green). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

event and to the amount (number or mass) of the scavenged material, (Hales et al., 1988).

During precipitation events of several hours certain air mass changes are inevitable, which may affect any scavenging conclusions. As a rough means of compensating for changes in the input aerosol during events we utilized a measured air pollutant that we assumed to be unaffected by sub-cloud scavenging. NO_x is measured at all three stations and has very low Henry's law constant for water as solvent, (Sander, 2015). Thus, we used the ratio of average NO_x concentrations during the pre-event periods to average NO_x -concentrations during the events to compensate for aerosol changes caused by varying air mass.

Before we discuss size dependent scavenging we present in Fig. 9 sub-cloud scavenging coefficients for the integral particle properties total number (NTO, cm^{-3}), total volume, (VTO, $\mu\text{m}^3\text{dm}^{-3}$), PM10 mass concentration, and black carbon, (BC, ngm^{-3}). Within 20% the scavenging coefficients of the three parameters agreed.

The ranking order of scavenging of NTO, VTO, and PM10 at all stations is the same, with NTO exhibiting lowest and PM10 the highest sub-cloud scavenging. The highest scavenging of PM10 we explained by this parameter encompassing mass contributions beyond the upper size limit of the MPSS, (800 nm), for which models predict steeply rising scavenging coefficients, (Slinn, 1977; Volken and Schumann, 1993). BC lies in between number and volume scavenging. The result for BC was somewhat counter-intuitive because one might expect BC to be

Fig. 9. Sub-cloud scavenging coefficients, (10^{-5}s^{-1}), of total number, (NTO), total volume, (VTO), Particulate mass $< 10\mu\text{m}$ particle diameter, (PM10), and black carbon, (BC), averaged over all precipitation events at the three GUAN stations Melpitz, Neuglobsow, and Waldhof. The error bars give ± 1 standard deviation of the mean values. PM10 was only available at Neuglobsow and Waldhof.

Fig. 10. Average size-dependent sub-cloud scavenging coefficients, (10^{-5}s^{-1}), at the three GUAN stations Melpitz, (ME, blue), Neuglobsow, (NG, red), and Waldhof, (WA, green). The grand average over all stations, (GA), is drawn in black. The error bars give ± 1 standard deviation of the mean values. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

scavenged less because of its (initially) hydrophobic character, (e.g., Covert and Heintzenberg, 1984; Hallberg et al., 1994). However, averaged over all events and stations $\Lambda_{\text{BC}} = 3.5 \cdot 10^{-5} \text{s}^{-1}$, which is more than twice as high as the value of $1.64 \cdot 10^{-5} \text{s}^{-1}$, reported by Latha et al. (2005) for urban conditions in Hyderabad, India. BC-concentrations at the GUAN-stations ranged between the detection limit of the MAAP (Petzold et al., 2002, $\approx 100 \text{ngm}^{-3}$), and $20 \mu\text{gm}^{-3}$ with a median value around 500ngm^{-3} . Latha et al. (2005) reported a range of $4\text{--}88 \mu\text{gm}^{-3}$ for their BC-concentrations. We speculated that the BC in the non-urban environment of the three GUAN-stations may have been somewhat aged and less hydrophobic compared to that in an urban source region such as Hyderabad.

General shape and absolute values of the size-dependent sub-cloud scavenging coefficients in Fig. 10 are similar to the results given by Laakso et al. (2003) for six years of data taken at the Finnish background station Hyytiälä. Their size range of ≈ 20 to $\approx 500 \text{nm}$ was somewhat smaller than that of the present study.

Depending to some extent on rain intensity their results scatter by more than a factor of five and show increasing scavenging coefficients below a minimum around 200nm , and a less steep increase towards larger sizes, which qualitatively agrees with other results, (Slinn, 1977; Volken and Schumann, 1993). However, others did not report the consistent structure that our results show at all three stations with local minima in scavenging around 60 and 200nm , and a weak local maximum around 120nm . Excepting identical instrumental problems at all stations, we can only speculate about reasons for the structure in our size-dependent scavenging results. They might have been affected by the modal structure of the PNSDs and by systematic differences in the physico-chemical properties of the different modes of the distribution.

Laakso et al. (2003) found a rather strong dependency of their median scavenging coefficient, Λ_{median} , over the range $20\text{--}510 \text{nm}$ on the rain rate, with a factor of two or more increase of Λ_{median} from 0.5 to $\approx 4 \text{mmh}^{-1}$. In Fig. 11 we show Λ_{median} , over the range $10\text{--}800 \text{nm}$ of the present study taken over all three stations, which indicated a somewhat smaller dependency of sub-cloud scavenging on rain rate. With the rain rate increasing from 0.5 to $\approx 4 \text{mmh}^{-1}$ Λ_{median} increased only by a factor of 1.4 , albeit starting at a higher level of scavenging than in the Finnish study.

3.2.2.2. Inter-station scavenging. Precipitation along trajectories connecting the three GUAN stations was utilized to study potential aerosol particle scavenging during aerosol transport on a scale of 200km . Most suitable for this study is the path $\text{WA} \rightarrow \text{NG}$ because

Fig. 11. Median sub-cloud scavenging coefficients, (10^{-5}s^{-1}), taken at all three stations over the diameter range 10–800 nm in five classes of rain rate per event indicated by horizontal error bars. Vertical error bars indicate 25% and 75% percentiles of the scavenging coefficients.

here by far the highest number connected data hours were available, combined with relatively few aerosol sources on the path, (cf. Table 2), and favorable meteorological conditions, i.e. frequent precipitation along the path. The other two paths in regime two had far less numbers of connected hours and much less favorable meteorological conditions, i.e. there were only few precipitation events along the paths ME → NG, and ME → WA.

As in the case of inter-station particle growth a simple model is formulated to describe the PNSD at the end of the transport path at station B with the PNSD at the starting point, (station A). Simply applying the scavenging coefficients derived at the individual stations at precipitation events occurring between A and B was not sufficient to describe the connected results at station B. As an example we show in Fig. 12 average PNSDs on the path WA → ME for 613 events of ≥ 1 mm precipitation during at least three transport hours. Just wet-scavenging PNSDs at station WA yielded much too low number concentrations at station ME. Even optimizing the level of the applied wet scavenging coefficients with Microsoft Excel's® GRG-solver left substantial discrepancies between the scavenged WA-PNSD and the ME-PNSD, (cf. curve optimized WAscav in Fig. 12). Consequently, we had to assume some aerosol sources along the path, which we simulated with two lognormal distributions.

Fig. 12. Average particle number size distributions before, (WA_start, green), and after, (ME_end, blue), 613 transport events with precipitation ≥ 1 mm for at least three hours. The dotted green line indicates the potential size distribution at station ME if only wet scavenging of the starting aerosol had occurred. The dotted black line gives the optimized scavenged size distribution of station WA. The red line indicates the wet-scavenged size distribution of station WA augmented with two optimized lognormal distributions representing the aerosol sources along the transport. Vertical error bars show two standard deviations of the mean values. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Fig. 13. Median sub-cloud scavenging ratios in seven classes of precipitation sums, (SRR, mm) indicated by horizontal error bars, along the transport path from Waldhof to Melpitz, WA → ME, and Neuglobsow to Melpitz, NG → ME. Vertical error bars indicate 25% and 75% percentiles of the scavenging ratios.

The six parameters of the two lognormal distributions were optimized with Microsoft Excel's® GRG-solver so that their sum plus the sub-cloud scavenged data from station WA fit the data at station ME within a few percent. For the example in Fig. 12 the parameters were added to Table 2.

Following, e.g., Tomasi et al. (2017), we defined a sub-cloud wet scavenging ratio as the concentration ratio of an air pollutant during or at the end of a precipitation event to its value before the event. This ratio describes the integral effect of precipitation after air mass transport between two stations. Accordingly, scavenging ratios were derived by subtracting the two lognormal distributions representing the on-path particle sources from the measured PNSD at station B and dividing the residuum by the un-scavenged results at station A.

Median scavenging ratios were calculated over the central part of the size range of the particle spectrometers, (20–200 nm), and displayed in Fig. 13 as a function of precipitation sums along the transport paths WA → ME and NG → ME.

Over the range of 0.1 to ≈ 50 mm of precipitation along the transport path median scavenging ratios changed by a factor of ≈ 1.9 . Similar to the scavenging coefficients in Fig. 11 they planed out at higher rain sums. Within the 25%–75% percentiles the scavenging coefficients of the two transport paths agreed. Whereas in light precipitation, (≤ 1 mm) more than 80% of the initial aerosol particles in the considered range survived the transport over ≈ 200 km more than 50% was scavenged by more than 15 mm of precipitation during aerosol transport.

4. Summary and conclusions

The present study was based on hourly aerosol particle and ancillary data taken at three GUAN stations in Northeastern Germany 150–200 km apart in the years 2009 through 2015. The aim was to investigate systematic process related differences that might show up when connecting their data with forward and backward air mass trajectories. The analysis of aerosol changes during air mass transport between the stations focused on the two low-pollution-to-high-pollution pathways of similar distance around 200 km from Waldhof, (WA) to Melpitz, (ME) and Neuglobsow, (NG), to Melpitz. Despite rather different initial PNSDs the increases in total concentration of particle number, (ΔNTO), volume, (ΔVTO), and BC, (ΔBC), agreed within 15%. Systematic variations were found in ΔNTO , ΔN_{26} , ΔVTO , and ΔBC as a function of time of day at which an air mass transport from WA to ME took place that could be explained with related source and transport processes.

With Radar-based precipitation data at the stations and along the

connecting trajectories sub-cloud particle scavenging was investigated at and in between the stations. At each station sub-cloud particle scavenging coefficients were determined as a function of particle size between 10 and 700 nm diameter, and as a function of precipitation sums. Median particle scavenging coefficients taken over the central part of the size range of the study strongly increased between 0.2 and 1 mm of precipitation and then planed out towards higher precipitation values. With the particle scavenging coefficients determined at the individual stations and precipitation data along connecting trajectories the sub-cloud particle scavenging analysis was extended to wet scavenging along the paths WA → ME, and NG → ME yielding median sub-cloud scavenging ratios over the range 0.2–25 mm of precipitation sums with a non-linear shape similar to that of the median particle scavenging coefficients at the individual stations. Whereas in light precipitation more than 80% of the initial particles in the considered range survived 150–200 km of transport, more than 50% was scavenged by more than 15 mm of precipitation during aerosol transport.

The present study used rather simplistic approaches to extrapolating aerosol data from a network of stations and to describe and quantify the effects of dry and wet aerosol processes between distant stations. It yielded new insights into processes leading to aerosol transformations. The accumulating GUAN data base however, offers much more than that with more detailed studies involving complex chemical/physical aerosol dynamics model such as presented by Tunved et al. (2010), for which the GUAN data base could be a test bed and with which more details of aerosol processes over the European region could be elucidated.

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