Rev. 1

Comment:
The methodical part of the back trajectory analysis is explained in section 3. There is, however, no description how exactly the back trajectories are used and how the maps are calculated. This methodical chapter must be much more detailed (and might also include the most important equations) so that the reader can reconstruct what exactly has been done. This is necessary for understanding the meaning, limitations and the interpretation of the maps as given by the authors later in the paper. Some examples: It remains unclear how the authors treated back trajectories that did not interact with the planetary boundary layer. How exactly were source regions defined?
Response: The revised text has been greatly expanded in response to the criticism by both reviewers. We refer to specific answers to the corresponding questions by rev. 2.

Comment:
On page 10 the authors write that the calculated immission maps have little in common with the emission data as shown in Fig. 2. Later in lines 239 and 240 it is said that "dilution through mixing with cleaner air masses and wet scavenging through in-cloud and sub-cloud processes" are two major atmospheric processes during transport. This is certainly true, it remains however unclear how such processes are incorporated in the back trajectory analysis. As the paper is now, I have the impression that the calculated immission maps are largely dominated by such atmospheric processes and the interpretation in terms of potential source regions is difficult. The signal from the potential source regions seem to be masked by such processes, e.g. strong dilution of emissions during westerly winds.
Response: This comment rightly touches one of the key points of our paper, namely our results that illustrate that emission maps not necessarily can be equated with the distribution of effect-related **immissions** in given receptor areas.
To clarify the information about HYPLIT4 we added to the model introduction: "Turbulent atmospheric mixing is included in parameterized form in HYSPLIT4. The present study utilizes the default version of this parameterization according to Draxler and Hess (1998). The back trajectories are calculated with the base version of HYSPLIT4 that does not include any specific dispersion and scavenging of atmospheric trace substances."

Comment:
For the second approach (connection of emissions and aerosol observations), an optimization algorithm was applied. There is again hardly any information what exactly has been done, except that the name of the used solver is given. This should be changed, please explain the applied optimization method in more detail. Maybe provide the relevant equations.
Response:
We greatly enlarged the respective text, including two equations:
. We expected both, seasonal variations and a long-term trend in the emissions. For M hours per month of measured components at the four stations the annual average EDGAR-emissions E_{PM10}, E_{BC}, E_{SO2}, and E_{NOx} were summed up along the 121 trajectory steps leading to the stations. Then monthly medians $\tilde{E}_{i=1,4}$ were formed according to Eq. 1 (exemplified for BC). Medians were chosen to reduce the effect of outliers due to local emission and scavenging events.

$$\tilde{E}_{BC} = Median(\sum_{n=1}^{121} E_{BC})_{m=1,M} \quad \text{Eq. 1}$$

The monthly median emission sums $\tilde{E}_{i=1,4}$ were modified with a monthly (f_m) and an annual factor (g_y) in order to simulate respective median monthly measured concentrations taken over all stations. Thus, for each component 12 monthly and 10 annual trend factors determined the agreement of modified summed emissions and measured concentrations. As objective or utility function χ^2 the sum of squared deviations between annually and monthly modified emission sums and monthly median measured concentrations was formed taken over the 120 months of the present study (exemplified for BC in Eq. 2).

$$\chi^2_{BC} = \sum_{j=1}^{120} \left(f_{m=1,12} \cdot g_{y=1,10} \cdot \tilde{E}_{BC} - eBC\right)^2 \quad \text{Eq. 2}$$

χ^2 was minimized with a Generalized Reduced Gradient (GRG) solver (Lasdon et al., 1978) that optimized the12 monthly and 10 annual factors for each of the four measured components. We used Excel's® implementation of the GRG solver procedure for the optimization. After optimization of month and trend factors the average relative deviation between emission-simulated and measured monthly median curves is 14%, 21%, 25%, and 18% for PM_{10}, eBC, SO_2, and NO_x, and respectively

Comment:
Page 7, lines 139-141: The authors say that equivalent back carbon was measured using a MAAP and converting the measured absorption signal into black carbon by using the default mass absorption cross section of the instrument (6.6 m2g-1). For using the terminology of equivalent black carbon (eBC) it is to my knowledge required that the mass absorption cross section is determined specifically for the site of interest, e.g. from parallel measurements with elemental carbon (EC) as determined using an analytical method such as the thermal optical method (see cited paper by Petzold et al 2013). The authors should therefore not call their measurements equivalent black carbon (eBC) but instead black carbon (BC)
Response:
In the instrumental section we added the two references (Birmili et al., 2016; Nordmann et al., 2013) that give details on the ways that the MAAP data at the GUAN-stations are validated against two analytical methods (Raman spectroscopy and thermal analysis) as requested by the reviewer. Consequently, we would like to maintain our use of term eBC.

Comment:

On page 8, line 172 the authors state that "Precipitation along the trajectories was used in the interpretation of the immission maps". How has this been done?
Response:
An explanation was given in the next sentence of the original manuscript, obviously not enough. Thus, we enlarged the respective text to:
Precipitation along the trajectories was used in the interpretation of the immission maps. The precipitation values mapped in the present study are those listed by HYSPLIT4 at each point of a trajectory. They are precipitation rates at the nearest grid cell of the assimilated global meteorological fields provided by the U.S. National Weather Service's National Centers for Environmental Prediction (NCEP) (Kanamitsu, 1989).

Comment:
Page 13, lines 298 – 300. The authors write that contributions from wind erosion of agricultural soils are not incorporated in present anthropogenic inventories. What about anthropogenic precursor emissions of secondary PM? How are they included or not included in this study. The authors should provide the corresponding discussion and information.
Response: The EDGAR data base utilized in the present study covers primary emissions only. Thus, our discussion is limited accordingly. In the revised text of the introduction of the EDGAR data base this has been clarified.

Comment:
Page 28, Table 2. It is surprising that the calculated percentage change in PM10 emissions relative to 2009 is highly sensitive to the considered time period (for 2009 – 2017 percentage change is -16%, for 2009 – 2018 percentage change is -6%). This deserves some explanation: Can this be a matter of limited robustness of the applied method, what are the uncertainties of the trend estimations?
Response: This change in percentage change is due to the general increase in emissions after 2017 that is clearly seen in Fig. 5 and in recent preliminary data shown by the European Environmental Agency (e.g., https://www.eea.europa.eu/data-and-maps/dashboards/air-quality-statistics-expert-viewer). Concerning the uncertainties of the trend estimations we added error bars to the trend figures. As estimates of the uncertainties these error bars were calculated by averaging the 12 monthly relative differences between measured and emission-simulated measurements for each year and applying these annual average relative differences as relative uncertainties to the calculated trends.

Comment:
Page 7, lines 157-158: "Through combustion processes the trace gases NOx and SO2 are related to anthropogenic aerosol formation". This is not correct or at least unclear. Of course, NOx and SO2 are in Germany mainly emitted from anthropogenic sources and processed in the atmosphere to form aerosols. However, the sentence should be re-phrased to something like "NOx and SO2 emitted by anthropogenic combustion processes are transformed in the atmosphere and add to the anthropogenic aerosol".
Response: Text changed as requested.

Comment:
Page 9, line 204: What is the "Southeastern half of the map"? Please be more accurate.
Response: Sorry, but we do not really understand the reviewer's problem here. The complete incriminated sentence reads "Highest average concentrations are measured in 204 airmasses from the Southeastern half of the map, most strongly expressed in PM_{10} and eBC 205 with maxima in a region leading from Southwest Poland through the Czech Republic, Slovakia,

206 Austria, and former Yugoslavia to Northeastern Italy, "i.e., the countries meant by „Southeastern half of the map" are specified.

Comment:
Page 9, line 207: Typo, "PM10and". Response: Typo corrected.

Comment:
Page 9, line 212: It is unclear what countries/ region is meant here? Rephrase so that this is clear,
Response: The incriminated part of the sentence reads: "…the emissions in the former countries stayed nearly constant...". We used the term „former" because we did not want to burden the reader by repeating the list of countries specified 3 sentences before but repeated it now in the revised text: „the emissions in Poland, Czech Republic, Slovakia, Austria, former Yugoslavia, and Italy stayed nearly constant or even increased in recent years"

Comment:
Page 12, line 279: Delete "are". Response: Done

Comment:
Page 12, line 288: I cannot see an increase in PM10 in 2010, neither in Fig. 4 nor in Fig. 5. Please explain or revise.
Response: The original text was unclear. We revised it to: "In 2010/2011 the trend curves of PM_{10} and NO_x in Fig. 5 show a slight increase that can be linked to a recovery of economic activity after the world-wide financial and economic crisis during the period 2007-2009. The increase in PM_{10} is also visible in the trend curves relative to 2005 published by the German Environment Agency (https://www.umweltbundesamt.de/daten/luft/luftschadstoff-emissionen-in-deutschland/emissionen-prioritaerer-luftschadstoffe).

Rev. 2

Comment:
For completeness for a reader not that familiar with this topic, it would nice to have at least one paragraph shortly summarizing whether similar work has been done outside Europe, for example in Norther America or Asia.
Comment: Outside Europe we only succeeded to identify two comparable studies in China for which we added the text: "With a much larger data set spanning a much tighter network of 1500 stations in China Rohde and Muller (2015) used the Kriging interpolation approach (Krige, 1951) to construct air pollution maps over China. Another approach to construct pollution maps over the province Henan, China was used by Liu et al., (2018). They combined an emission inventory with chemical modeling and back trajectories to derive high resolution maps of particulate and gaseous pollution components."

Comment:
First, the paper does not report anything about data coverage or its quality. Were there any major data gaps, not apparent in Table 1, that might influence the analysis performed in the paper?
Response: Table 1 has been augmented with the number of validated data hours for each measured component and the text related to Table 1 has been extended to: "Table 1 gives an overview over the instrumental characteristics of all stations and the total number of validated data hours for each utilized component. The minimum is 57962 hours for validated SMPS-data at the tree GUAN-stations and the maximum with 88838 validated data hours for NO_x at all four stations. Strictly concurrent (by the hour) are less validated data hours. For SMPS, eBC, and SO_2-data at the GUAN-stations this numbers is 48533 hours, and 48114 and 47729 hours for PM_{10} and NO_x-data, respectively, at all four stations. However, these reduced strictly concurrent numbers do not substantially affect the 10-year-average maps discussed below."

Comment:
Is all the data quality checked and how? Do the detection limits of different instruments, especially what it comes to trace gas data, play any role here?
Response: The text on data quality has been greatly expanded and reads now as "TROPOS-type mobility particle size spectrometers (MPSS, Wiedensohler et al., 2012) were used to record particle number size distributions across the particle size range 10-800 nm. Quality assurance of the long-term measurements followed the recommendations of Wiedensohler et al. (2018) including weekly inspections as well as monthly and annual maintenance intervals. Once a year the MPSS were intercompared against a reference MPSS of the WCCAP (World Calibration Center for Aerosol Physics) either on-site and/or at the calibration facility. The lower detection limit of the MPSS is around 100 cm-3 for a time resolution of 5 minutes. Equivalent Black Carbon (eBC) was determined by multi-angle absorption photometers (MAAP) using a mass absorption cross section of 6.6 m^2 g-1 (Petzold et al., 2013; Nordmann et al., 2009; Birmili et al., 2009). An intercomparison of multiple MAAP instruments resulted in an inter-device variability of less than 5% (Müller et al., 2011). While the MAAP deployed at the TROPOS station Melpitz was biannually compared to the reference absorption photometer at the WCCAP in Leipzig, the instruments at the UBA stations Waldhof and Neuglobsow were serviced by the manufacturer. For hourly measurements of PM10 continuous oscillating microbalances (Thermo Scientific TEOM 1400) were utilized at stations CO, NG, and WA. At station ME PM10 data were determined in daily filter samples (0:00 to 24:00 CET), Spindler et al. (2013). The TEOM1400-instrument and gravimetric filter sampling are different methods for particle mass concentrations. The TEOM collects particulate mass on a vibrating substrate (tapered element) and registers the change of the oscillation frequency that

is decreasing with mass loading (Patashnick and Rupprecht, 1991). The TEOM operates at a constant temperature setting above ambient (typically 30– 50°C) to prevent contraction and expansion of the tapered element and reduce interferences from water vapor condensation. However, heating the ambient air enhances volatilization of particle-bound semivolatile compounds (e.g., ammonium nitrate and some organic species) resulting in an underestimation of PM when semivolatile dominate the particulate phase during cold seasons. The condensation and evaporation of ammonium nitrate and organic species can also influence the filter sampling under ambient conditions. Here the effect can be balanced partly by the temperature variation during the daily filter sampling. However, the results of both methods mostly are in good agreement (e.g., Zhu et al., 2007)….At the three GUAN stations both are measured with the same temporal resolutions as the aerosol data. Additionally, at Collmberg NOx-data could be utilized in the interpretation of the aerosol data. The trace gas analyzers for NOx and SO2 were calibrated with test gases for NO (NO in N2) and SO2 (SO2 in N2, both Air Liquide, Germany). NO2 was produced in a gas phase titration device (GPT APMC370, Horiba, Germany) by quantitative oxidation of NO test gas (Rehme, 1976). The trace gas analyzers were used in an optimal range and all registered values (also below the detection limit) were used for this long-term study.

Comment:
Second, there are no continuous PM10 data for Melpitz, neither SO2 data for Collmberg. How does this influence the analysis of this paper?
Response: Basically, eliminating one of the stations from the maps only reduces the statistical solidity in some of the geocells but not the overall geographic patterns. In particular the continuous PM10-data at Collmberg located only 30 km away from Melpitz supports the daily average PM10-data at Melpitz. We added the following text and two references stating that TEOM1400 and gravimetric filter sampling are different methods but are mostly in good agreement.
"The TEOM1400-instrument and gravimetric filter sampling are different methods for particle mass concentrations. The TEOM collects particulate mass on a vibrating substrate (tapered element) and registers the change of the oscillation frequency that is decreasing with mass loading (Patashnick and Rupprecht, 1991). The TEOM operates at a constant temperature setting above ambient (typically 30– 50°C) to prevent contraction and expansion of the tapered element and reduce interferences from water vapor condensation. However, heating the ambient air enhances volatilization of particle-bound semivolatile compounds (e.g., ammonium nitrate and some organic species) resulting in an underestimation of PM when semivolatile dominate the particulate phase during cold seasons. The condensation and evaporation of ammonium nitrate and organic species can also influence the filter sampling under ambient conditions. Here the effect can be balanced partly by the temperature variation during the daily filter sampling. However, the results of both methods mostly are in good agreement (e.g., Zhu et al., 2007)."

For the measurements described in this paper there are also results from a successful one-year intercomparison of TEOM1400 and filter sampling in winter 2012/13 in the region (city of Dresden) of special interest. Results available at https://www.luft.sachsen.de/download/luft/Vergleichsmessungen_PM2_5_Internet.pdf
The highest time resolution discussed in the present study is one month while most results are discussed as 10-year averages. Thus, utilizing daily PM_{10}-averages from the Melpitz station did not affect the results significantly.

Comment:
Third, what is purpose of presenting PM0.8 derived from SMPS if that is not used anywhere in the paper?

Response: Sorry about that. This was just a typo that had not been eliminated from a previous version of the manuscript.

Comment:
One final issue: Figure 1 caption refers to particle volume concentration when showing PM10.
Again, sorry about that. This was just a typo that had not been eliminated from a previous version of the manuscript.

Comment:
What is the benefit of displaying particle number concentrations plots (Figs. 1, top and bottom left) for the rest of this paper? Particle number concentrations are not related to any of the discussions about emissions later in the paper. The authors briefly mention atmosphere new particle formation as one particle source, and emphasizes SO2 plumes or lofted layers as important locations for this source. If these two sources dominated new particle formation over Europe, how would they explain the common observation of regional new particle formation events, taking simultaneously place over tens to hundreds of km scales, in a vast number of surface measurement sites in Europe.
Response: Thank you for criticizing the lack of motivation and limited discussion of particle-number related results. First, we eliminated the discussion of $N_{10\text{-}26}$ because of the more locally controlled nature of this parameter. Then we expanded the discussion of $N_{10\text{-}800}$ substantially with seasonal maps and a trend discussion connected to gas phase and meteorological parameters.

Comment:
As mentioned by the authors, PM10 in Figs. 1 and 2 have practically no resemblance with each other. Noting that both these data sources are used together in later analyses, the authors should discuss this issue and its potential consequences in their analysis. What are the potential reasons for the differences between these two figures? Why the map in Figure 1 does not capture the emission hotspots of Figure 2? There is strong PM10 gradient from North-West to South-East in Figure 1. Why does Figure 2 not give any indication about such gradient? Are the main PM10 in the South-East direction located outside the map area?
Response: In the section on emissions we first qualified the emission data by "Primary aerosol emission data are generally characterized by rather high uncertainties. For the EDGAR data base Crippa et al. (2018) report a range of variation in 2012 between 57.4% and 109.1% for PM_{10}, and between 46.8% and 92% for BC. Even higher uncertainties in PM emissions might come from super-emitting vehicles that are not considered in this data base (Klimont et al., 2017)." We also added extensive text and a new figure to the manuscript that resolve the seeming discrepancy between immission and emission maps. Additional to the original manuscript the validity of the present approach of connecting immission and emission of particulate pollution was tested by calculating temporal changes of eBC for subsets of back trajectories passing over two separate prominent emission regions, region A to the Northwest and B to the Southeast of the measuring stations. Consistent with reported emission data the calculated immission decreases over region A are significantly stronger than over region B.

Comment:
The authors state that NOx and SO2 maps look very similar (related to figures 1 and 2). I wonder why these results are shown in the paper, especially when considering that both these trace gases are important parts of trend analyses presented later in the paper.
Response: In a new supplement to the manuscript we display seasonal maps of ozone SO_2, and NO_x.

Comment:
Based on Figure 3, the authors discuss the influence of wind speed and wet scavenging on measured concentrations. Unfortunately, this discussion remains very qualitative. If included in this paper, more concrete results on the influence of these two variables should be presented.
Response: We agree with the reviewer concerning the qualitative nature of the criticized discussion. It is due to the fact that the HYSPLIT trajectory model only provides very limited meteorological information along the trajectories taken from neighboring grid-points of the used meteorological maps. Nevertheless, we feel that the maps provided in Fig. 3 provide a useful illustration of possible reasons for the discrepancies between the emission and the immission maps.

Comment:
In the last part of the paper, the authors investigate the time evolution of emissions during the past 10 years. I have a couple of concerns related to this. First, the authors only stated that they optimized the annual and monthly emissions of 2009 for the rest of the time series using their measurements. This is too scarce information. The authors should provide more details on how this exercise what done in practice.
Response: See response to same comment by reviewer 1

Comment:
Second, as well known, emission changes have been very different in different parts of Europe during the past years. Is the approach applied in this paper able to catch this feature in any way, and if it is not, this should be explicitly mentioned in the paper.
Response: In the subsection of lines 294 through 322 of the original manuscript we take up this issue, extending our discussion of possible influences from neighboring countries all way to Romania.

Comment:
The final conclusion of this paper remain somewhat vague. The authors should more concrete summarize what new information this study brings on top of what is already known about the past emission changes in Europe. The authors mention some of the limitations of the current study yet, based on the comments give above, I feel that this list could be expanded a bit.
Response: More specific information was added to the conclusions and the list of limitations of the study was expanded.

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Aerosol immission maps and trends over Germany with hourly data at four rural background stations from 2009 to 2018

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Abstract

Ten years of hourly aerosol and gas data at four rural German stations have been combined with hourly back trajectories to the stations and inventories of the European EDGAR emission database yielding immission maps over Germany of PM_{10}, particle number concentrations, and equivalent black carbon (eBC). The maps reflect aerosol emissions modified with atmospheric processes during transport between sources and receptor sites. Compared to emission maps strong Western European emission centers do not dominate the downwind concentrations because their emissions are reduced by atmospheric processes on the way to the receptor area. PM_{10}, eBC, and to some extent also particle number concentrations are rather controlled by emissions from Southeastern Europe from which pollution transport often occurs under dryer conditions. Newly formed particles are found in air masses from a broad sector reaching from Southern Germany to Western Europe which we explain with gaseous particle precursors coming with little wet scavenging from this region.

Annual emissions for 2009 of PM_{10}, BC, SO_2, and NO_x were accumulated along each trajectory and compared with the corresponding measured time series. The agreement of each pair of time series was optimized by varying monthly factors and annual factors on the 2009

Gelöscht:

Gelöscht: necessarily

emissions. This approach yielded broader summer emission minima than published values that were partly displaced from the midsummer positions. The validity of connecting immission and emission of particulate pollution was tested by calculating temporal changes of eBC for subsets of back trajectories passing over two separate prominent emission regions, region A to the Northwest and B to the Southeast of the measuring stations. Consistent with reported emission data the calculated immission decreases over region A are significantly stronger than over region B.

Gelöscht: For BC, SO_2, and NO_x stronger emission-reductions were determined than what German and European environmental agencies reported. These findings are emphasized with 2017 as endpoint of the trend from which on our study shows emission increases. Comparing calculated trends with emission trends in neighboring countries as published by EEA supports the explanation that the observed trends are to some extent due to changes in imported air masses. Most strongly this holds for SO_2, the trend of which follows that of Romanian emissions rather well.

1 Introduction

The atmospheric aerosol is known to influence the Earth's radiation budget because it directly scatters and absorbs solar radiation (Schwartz, 1996; Bond et al., 2013), and acts as cloud condensation nuclei, thus modulating the optical properties and lifetimes of clouds (Twomey, 1974; Penner et al., 2004). In many regions of the globe that had undergone industrialization early on, anthropogenic aerosol concentrations are currently in decline (Leibensperger et al., 2012; Zanatta et al., 2016). With respect to declining concentrations and emissions, Samset al. (2018) suggest that removing present-day anthropogenic aerosol emissions – assuming constant greenhouse gas emissions, could lead to a global mean surface heating as high as 0.5–1.1°C.

Besides climate, the atmospheric aerosol has been acknowledged to influence human health through respiratory and cardiovascular health endpoints (Anderson et al., 2012). Lelieveld et al., (2015) quantified the world-wide burden of disease (premature mortality) due to outdoor pollution, large part of which was attributed to airborne particulate matter. It is apparent that the distribution of adverse health effects is very uneven among the world-wide population, depending on the local level of outdoor pollution.

In view of the described man-driven effects it seems imperative to develop instruments to reliably monitor changes in anthropogenic aerosol concentrations as well as an understanding of the balance between aerosol sources and measured concentrations. Researchers have strived to obtain a spatial picture of the distribution of pollutants, and to achieve a connection between the sources of pollution and concentrations downwind. A widely used method has been the extrapolation of concentrations measured in one or several locations into two-dimensional space through the use of meteorological dispersion approaches: The first maps of particulate

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73 air pollutants over Europe were constructed in the 1970s with the help of coarse emission data
74 and simple trajectory models (Eliassen, 1978). Statistical methods were developed to connect
75 pollution sources and ensuing aerosol concentrations at receptor sites (Miller et al., 1972;
76 Friedlander, 1973; Cass and McRae, 1983). By combining statistics with back trajectory data
77 sectorial information about sources controlling the composition of the aerosol over Southern
78 Sweden was derived by Swietlicki et al., (1988). Later the approach of using back trajectories
79 to map aerosol sources was refined by Stohl (1996) and tested with one-year sulfate data from
80 the co-operative program for monitoring and evaluation of the long-range transmission of air
81 pollutants in Europe (EMEP, www.emep.int). In a similar approach with five years of aerosol
82 data from a single Siberian receptor site Heintzenberg et al. (2013) identified potential source
83 regions over Eurasia and with aerosol data from four Swedish icebreaker expeditions over the
84 Central Arctic (Heintzenberg et al., 2015). Charron et al. (2008) constructed concentration field
85 maps to identify the source regions of specific types of aerosol particle size distributions
86 arriving in England. All these works share the approach that time-dependent information on
87 concentrations measured at receptor site(s) are transformed into space, thus allowing
88 conclusions on the potential source regions of gaseous and/or particulate emissions.
89
90 With more comprehensive air quality models concentrations of specific aerosol were
91 mapped over Europe together with short temporal developments (e.g., Schell et al., 2001). For
92 specific episodes high spatial resolution aerosol concentration maps in urban and non-urban
93 European areas have been generated with sophisticated chemistry transport models (e.g.,
94 Beekmann et al., 2015; Riemer et al., 2004; Wolke et al., 2004). For the years 2002 and 2003
95 Marmer and Langman (2007) analyzed the spatial and temporal variability of the aerosol
96 distribution over Europe with a regional atmosphere-chemistry model. They found that
97 meteorological conditions play a major role in spatial and temporal variability in the European
98 aerosol burden distribution. Regionally, year to year variability of modeled monthly mean

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aerosol burden reached up to 100% because of different weather conditions.

In the present study ten years of hourly aerosol data at four German stations were available for the identification of potential source regions. As it appears unrealistic to analyze such a large database with advanced chemical transport models we resorted to the well proven approach of utilizing back trajectories cited above and connected the results to emission fields. We define the resulting concentration maps of particulate and gas parameters as immission maps because they represent long-term average emissions of air pollutants modified by the controlling atmospheric processes along the pathways to the receptor sites. In Charron et al. (2008) this approach is termed "concentration field map method". With a much larger data set spanning a much tighter network of 1500 stations Rohde and Muller (2015) used the Kriging interpolation approach (Krige, 1951) to construct air pollution maps over China. Another approach to construct pollution maps over the province Henan, China was used by Liu et al., (2018). They combined an emission inventory with chemical modeling and back trajectories to derive high resolution maps of particulate and gaseous pollution components and find that emissions from neighboring provinces are important contributors to local air pollution levels.

Recent political, economic and technological developments in Europe have caused substantial changes in the emission of air pollutants. Lavanchy et al. (1999) deduced a trend in atmospheric black carbon from preindustrial times to 1975. Strong downward trends in major aerosol components before and after the German reunification (1983-1998) over rural East Germany were reported by Spindler et al., (1999). For the years 2003 – 2009 Kuenen et al., (2014) published trends in the development of aerosol emissions as elaborated from reported emissions. The German Environmental Agency (GEA) publishes trends in air pollution as measured at a number of ca. 380 federal and state air quality stations (Minkos, 2019).

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According to these records, PM_{10} mass concentrations declined by approximately 25 % over the period 2000-2019

Combining long-term aerosol and gas data at the four stations of the present study provide an excellent data base for identifying both the most important source regions and possible temporal changes. During the ten recent years covered by our data we expected noticeable systematic changes in our time series that can be interpreted in terms of emissions. As a side result in the process of deriving long-term emission trends of major air pollutants over Germany information of the monthly disaggregation of annual aerosol emissions can be derived.

2 Aerosol and trace gas data

The core data of the present study have been measured at the stations Melpitz (ME), Neuglobsow (NG), and Waldhof (WA) of the German Ultrafine Aerosol Network GUAN network (Birmili et al., 2016) and at station Collmberg (CO) operated by the Saxonian Environment Agency. These four rural background stations lie in the northeastern lowlands of Germany at distances between 30 and 205 km from each other. Ten-year-average particle mass concentrations under 10 µm particle diameter (PM_{10}) and their standard deviations at the four stations are rather similar: 15±13, 22±12, 14±10, and 15±11 μgm^{-3} at CO, ME, NG, and WA, respectively. The corresponding long-term average particle number concentrations between 10 and 800 nm particle diameter (N_{10-800}) and their standard deviations at the three GUAN-stations are 5400±4100, 3600±2300, and 4300±2800 cm^{-3}, respectively. Basic statistics on particle number and eBC mass concentrations of the three GUAN-stations were presented in Sun et al. (2019) whereas details about instrumentation and their maintenance can be found in Birmili et

Gelöscht: Table 1 gives an overview over their characteristics.

al., (2016). The ensemble of hourly data at the four stations is the base of the pollution maps derived in this work.

TROPOS-type mobility particle size spectrometers (MPSS, Wiedensohler et al., 2012) were used to record particle number size distributions across the particle size range 10-800 nm. Quality assurance of the long-term measurements followed the recommendations of Wiedensohler et al. (2018) including weekly inspections as well as monthly and annual maintenance intervals. Once a year the MPSS were intercompared against a reference MPSS of the WCCAP (World Calibration Center for Aerosol Physics) either on-site and/or at the calibration facility. The lower detection limit of the MPSS is around 30 cm^{-3} for a time resolution of 30 minutes. Equivalent Black Carbon (eBC) was determined by multi-angle absorption photometers (MAAP) using a mass absorption cross section of 6.6 $m^2 g^{-1}$ (Petzold et al., 2013; Nordmann et al., 2013; Birmili et al., 2016). An intercomparison of multiple MAAP instruments resulted in an inter-device variability of less than 5% (Müller et al., 2011). While the MAAP deployed at the TROPOS station Melpitz was biannually compared to the reference absorption photometer at the WCCAP in Leipzig, the instruments at the UBA stations Waldhof and Neuglobsow were serviced by the manufacturer. For hourly measurements of PM_{10} continuous oscillating microbalances (Thermo Scientific TEOM 1400) were utilized at stations CO, NG, and WA. At station ME PM_{10} was determined in daily filter samples (0:00 to 24:00 CET), Spindler et al. (2013). The TEOM1400-instrument and gravimetric filter sampling are different methods for particle mass concentrations. The TEOM collects particulate mass on a vibrating substrate (tapered element) and registers the change of the oscillation frequency that is decreasing with mass loading (Patashnick and Rupprecht, 1991). The TEOM operates at a constant temperature setting above ambient (typically 30– 50°C) to prevent contraction and expansion of the tapered element and reduce interferences from water vapor condensation. However, heating the ambient air enhances volatilization of particle-

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bound semivolatile compounds (e.g., ammonium nitrate and some organic species) resulting in an underestimation of PM when semivolatile material dominates the particulate phase during cold seasons. The condensation and evaporation of ammonium nitrate and organic species can also influence the filter sampling under ambient conditions. Here the effect can be balanced partly by the temperature variation during the daily filter sampling. However, the results of both methods mostly are in good agreement (e.g., Zhu et al., 2007).

Hourly aerosol data from the three GUAN-stations during 2009 – 2015 (NG ≥2011) have been utilized in a previous study (Heintzenberg et al., 2018) to understand aerosol processes during air mass transport between the stations. In the present study the data set was enlarged to include the additional station Collmberg and data at all stations from the year 2016 through 2018. The integral aerosol parameters particle number concentration ($N_{10\text{-}800}$, cm^{-3}), light absorption-equivalent mass concentration of Black Carbon (eBC, μgm^{-3}), and particle mass concentrations under 10 µm particle diameter (PM_{10}, μgm^{-3}) were utilized. $N_{10\text{-}800}$ is based on the integral over measured particle size distributions from 10 to 800 nm.

NO_x and SO_2 emitted by anthropogenic combustion processes are transformed in the atmosphere and add to the anthropogenic aerosol. At the three GUAN stations both are measured with the same temporal resolutions as the aerosol data. Additionally, at Collmberg NO_x-data could be utilized in the interpretation of the aerosol data. The trace gas analyzers for NO_x and SO_2 were calibrated with test gases for NO (NO in N_2) and SO_2 (SO_2 in N_2, both Air Liquide, Germany). NO_2 was produced in a gas phase titration device (GPT APMC370, Horiba, Germany) by quantitative oxidation of NO test gas (Rehme, 1976). The trace gas analyzers were used in an optimal range and all registered values (also below the detection limit) were used for this long-term study. As most particle numbers in polluted continental environments tropospheric ozone is a secondary atmospheric pollutant. We utilized hourly

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Gelöscht: Of the total number of 87648 hours during the ten-year period 77516 hours with at least concurrent PM_{10}-data at all four stations could be utilized.

Gelöscht: number concentrations below 10 - 26 nm ($N_{10\text{-}26}$, cm^{-3}), and

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Gelöscht: Through combustion processes the trace gases NO_x and SO_2 are related to anthropogenic aerosol formation

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ozone data taken at all four stations throughout the studied time period as ancillary information in the discussion of particle-number related results. For the ozone measurements a common trace gas ozone monitor was used (Horiba APOA-350). This device quantifies tropospheric ozone by UV Absorption and use the cross-flow modulation principle. Ambient air with and without ozone (elimination by a selective scrubber) was used alternatively in the measuring cuvette yielding a very stable ozone signal. The devices were calibrated using an ozone-standard (Ozon-Calibrator, Thermo Environmental Instruments 49PS).

Table 1 gives an overview over the instrumental characteristics of all stations and the total number of validated data hours for each utilized component. The minimum is 57962 hours for validated MPSS-data at the three GUAN-stations and the maximum with 88838 validated data hours for NO_x at all four stations. Strictly concurrent (by the hour) are less validated data hours. For MPSS, eBC, and SO_2-data at the GUAN-stations this numbers is 48533 hours, and 48114 and 47729 hours for PM_{10} and NO_x-data, respectively, at all four stations. However, these reduced strictly concurrent numbers do not substantially affect the 10-year-average maps discussed below.

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3 Back trajectories

With the HYSPLIT4 model (Stein et al., 2015) and based on the meteorological fields from the Global Data Assimilation System with one-degree resolution (GDAS1, https://www.emc.ncep.noaa.gov/gmb/gdas/) three-dimensional trajectories were calculated arriving every hour at a height of 500m above ground level at the four stations. The trajectories were calculated backward for up to five days using the meteorological fields from the server at

Air Resources Laboratory (ARL), NOAA (http://ready.arl.noaa.gov), where more information about the GDAS dataset can be found. Turbulent atmospheric mixing is included in parameterized form in HYSPLIT4. The present study utilizes the default version of this parameterization according to Draxler and Hess (1998). The back trajectories are calculated with the base version of HYSPLIT4 that does not include any specific dispersion and scavenging of atmospheric trace substances. Precipitation along the trajectories was used in the interpretation of the immission maps. The precipitation values mapped in the present study and the temperature values used in the trend discussion of $N_{10\text{-}800}$ are those listed by HYSPLIT4 at each point of a trajectory. They are meteorological parameters at the nearest grid cell of the assimilated global meteorological fields provided by the U.S. National Weather Service's National Centers for Environmental Prediction (NCEP) (Kanamitsu, 1989). Average horizontal wind speeds in between two one-hour trajectory steps were calculated from the distance covered by a trajectory between two successive steps. With the 350593 hourly back trajectories from the four stations the time series of $N_{10\text{-}800}$, PM_{10}, and eBC were extrapolated over Germany and part of the neighbor countries. At Melpitz PM_{10}-data were only available as daily averages. Thus, the daily average concentrations were extrapolated along each hourly trajectory of the respective day.

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4 Emission data

For the interpretation of the immission maps we used the emission data set version 4.3.2 for 2009 of the components particle mass concentrations below 10 µm (PM_{10}), BC, NO_x and SO_2 as compiled in the Emissions Data Base for Global Atmospheric Research (EDGAR, https://edgar.jrc.ec.europa.eu/overview.php?v=432_AP, DOI (https://data.europa.eu/doi/10.29

04/JRC_DATASET_EDGAR). This data set concerns primary emissions only and has been introduced by Crippa et al., (2018). All human activities, except large scale biomass burning and land use, land-use change, and forestry are included in the data base. Emissions of coarse particles from agricultural surfaces are not included. They are, in fact, very sensitive to soil and weather conditions, and thus not trivial to quantify. Primary aerosol emission data are generally characterized by rather high uncertainties. For the EDGAR data base Crippa et al. (2018) report a range of variation in 2012 between 57.4% and 109.1% for PM_{10}, and between 46.8% and 92% for BC. Even higher uncertainties in PM emissions might come from super-emitting vehicles that are not considered in this data base (Klimont et al., 2017). In our maps and trend calculations we applied the grid values of emission data that were listed in the EDGAR inventories no more than 30 km away from any trajectory time step.

5 Results and discussion

5.1 Aerosol concentration maps (immission maps

The trajectory-extrapolated $N_{10\text{-}800}$, PM_{10}, and eBC from the four stations yielded immission maps averaged over the period 2009 – 2018, that are collected in Figs. 1-2. Both, the particle-number related $N_{10\text{-}800}$ and the particle-mass related PM_{10}, and eBC exhibit systematic seasonal variations. Most events of new particle formation (NPF) over the continents occur during the photochemically active summer months (Kulmala et al., 2004) whereas the particle-mass related aerosol parameters due to combustion processes exhibit highest concentrations during the winter months (Matthias et al., 2018). Consequently, we constructed two maps for each discussed component: One of averages over the months April through October and one of averages over the months November through March. Only map cells with at least 300 trajectory hits are discussed. Interpreting these hits in terms of Poisson-statistics would then yield a

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maximum uncertainty of 5.8% per cell. In terms of a Gaussian statistic the arithmetic cell-averages displayed in the maps exhibit standard deviations of cell averages that are less than six percent.

The maps of N_{10-800} in Fig. 1 show distributions of air masses over Germany and adjacent countries related to particle numbers instead of particulate mass. There are two arguments for showing maps of number related results. First, particle number concentrations are connected with cloud processes, their formation (Pruppacher and Klett, 1978), radiative effects, e.g., albedo (Twomey, 1974), and precipitation (Li et al., 2011). Second, in the area of aerosol-health issues ultrafine particles (< 100 nm diameter) have been gaining attention in recent years (Wichmann and Peters, 2000), i.e. an increasing number of health effects is attributed rather to particle number than to particle mass. The fact that NPF-events occur concurrently in or near the top of the continental planetary boundary layer over wide geographical regions (e.g., Wehner et al., 2007) is partly due to concurrent advantageous photochemical conditions allowing for the formation of condensable vapors, in particular global radiation (Birmili et al., 2001). Two other factors constraining NPF are the availability of gaseous particle-precursors and the concurrent pre-existing aerosol.

The summer map (4-10) of N_{10-800} exhibits the high values in the Southwest-to-Northeast-sector of the map. Highest values are concentrated in a belt reaching from Burgundy through Switzerland, Southern Germany, Czech Republic to Southwestern Poland. Interestingly, this belt of high N_{10-800} is collocated to large extent with a belt of high summer ozone concentrations (cf. Fig. S1). This photochemically controlled pollutant (Monks et al., 2015) exhibits highest summer concentrations in air masses from Southwestern Poland and Northern Czech Republic, a region from which high ozone values are reported (Struzewska and Jefimow, 2013; Hůnová, 2003; Hůnová and Bäumelt, 2018). However, the summer map of N_{10-800} does not show the

Gelöscht: (Kulmala et al., 2004)(Matthias et al., 2018)

Gelöscht: The highest map-coverage was reached with PM_{10} with at least 3951 data points in each geocell. As eBC and size distribution data were only available at the three GUAN-stations the derived maps contained a minimum of 751 data points per geocell.

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highest values in air masses from the region with highest ozone pollution. High particle numbers in air masses coming over the Alps from Northern Italy may be related to the high emissions of air pollutants in the Po Valley that are known to reach frequently through so called alpine pumping (Winkler et al., 2006; Lugauer and Winkler, 2005; Reitebuch et al., 2003) over the mountains. The high NO_x-concentrations in air masses from Northern Italy in both summer and winter maps (see Fig. S2) indicate that pollution from south of the Alps can even reach Northeastern Germany. In the winter map of N_{10-800} (11-3 in Fig. 1) the belt of highest summer values is apparently complemented by more transalpine pollution transport and by transport from the Southeast. The lower photochemical activity in winter is reflected in the lower winter ozone concentrations in Fig. S1, albeit transalpine pollution transport is still visible in the winter map of NO_x in Fig. S2. Northwestern Italy also shows up as an emission hot spot in the maps of trajectory-summed emissions in Fig. S4.

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In both summer and winter the maps of PM_{10}, and eBC in Fig. 2 exhibit a clear Northwest-to-Southeast structure with the cleanest sector being in the Northwest covering the coastal area of the North Sea, the BENELUX countries Belgium, the Netherlands, and Luxemburg, and Northwestern Germany. The strongest contrast between the cleanest Northwesterly and the most polluted Southeasterly map sectors is seen in the winter map of eBC. Highest average concentrations are measured in airmasses from the Southeastern half of the map, most strongly expressed in PM_{10} and eBC with maxima in a region leading from Southwest Poland through the Czech Republic, Slovakia, Austria, and former Yugoslavia to Northeastern Italy. The back trajectories in the Southeastern sector of the maps for PM_{10} and eBC point towards countries, in which the emissions of air pollution in the past 20 years developed very differently as compared to those in Western Europe. According to the European Environment Agency (https://www.eea.europa.eu/data-and-maps/dashboards/air-pollutant-emissions-data-viewer-2) the latter parts of Western Europe experienced a strong and nearly monotonous decrease in

emissions of PM_{10} whereas the emissions in Poland, Czech Republic, Slovakia, Austria, former Yugoslavia, and Italy stayed nearly constant or even increased in recent years after the dramatic decreases in the course of the political developments of the 1990ies. The seasonal maps of the combustion derived SO_2 in Fig. S3 look very similar to the those of the particle-mass related maps of PM_{10} and eBC, again the strongest NW/SE-contrast visible in winter.

5.2. Pollutant emissions and atmospheric processes

In Fig. 3 annual average emissions of PM_{10}, BC, SO_2, and NO_x are mapped for 2009 according to the EDGAR emission database. Except for the absolute numbers the maps for SO_2, and NO_x look rather similar to those for particulate emissions. They all emphasize highly populated and industrialized emissions center. Beyond that the SO_2-map accentuates individual large combustion sources such as conventional power plants. Whereas the strong emissions in Northern Italy are seen in the maps of PM_{10}, BC, and NO_x emissions in the countries in the Southeastern sector of the maps by no means reflect the high concentrations of particulate components seen in the immission maps of Figs. 1 and 2.

The seeming discrepancy between the immission maps in Figs. 1 and 2 and the emission maps of Fig. 3 can be resolved. For that purpose, the EDGAR-emissions of PM_{10}, BC, SO_2, and NO_x along all 350593 hourly back trajectories to the four stations during the ten studied years were summed up. Then the sums were extrapolated back along each trajectory. In Fig. S4 10-year average maps of these extrapolated emission sums are displayed. As in Fig. 3 except for the absolute numbers there is a strong similarity between the four mapped component sums. Because of the integral nature of the mapped results one cannot expect the maps in Fig. S4 to locate correctly specific emission centers. However, they certainly indicate the map sectors from which the most substantial emissions could have reached the stations. As in Figs. 1 and

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Gelöscht: →(Pruppacher and Klett, 1978)(Twomey, 1974)(Li et al., 2011)(Wichmann and Peters, 2000)(e.g., Wehner et al., 2007)In air masses from the extreme Southeastern sector of the map relatively low total number concentrations were measured. Even more so this holds for the relatively newly formed $N_{10\text{-}26}$ concentrations that exhibit a broad maximum in the Southwestern half of its map whereas $N_{10\text{-}800}$ has its maxima in a rather narrow band of air masses reaching from Switzerland through Southeastern Germany, Western Czech Republic to the former so called "Black Triangle" region near the Southeastern corner of Germany.¶
¶
→Current explanations of the new particle formation process (as indicated by $N_{10\text{-}26}$) point towards photochemical processes that take place in plumes that contain sulfur dioxide (Größ et al., 2018). Several authors have stressed the possibility of particles to be formed in lofted layers, which are subsequently mixed to the ground (Platis, 2016), and/or in sulfur-rich plumes downstream of industrial point sources such as power plants (Junkermann and Hacker, 2018). ¶
¶
→The trajectory extrapolated PM_{10}-concentrations in Fig. 1 most strongly show the contrast between the relatively clean Northwest sector and the high concentrations in the Southeast sector of the maps.

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2 the Southeastern sectors of the maps of integrated emissions most prominently show up. Interestingly, the maps in Fig. S4 also indicate the highly polluted region of Northwestern Italy (Diémoz et al., 2019a; Diémoz et al., 2019b). The emissions from the emission centers in Northwestern Europe are hardly discernible in Fig. S4. They do show up (most strongly in Fig. S4c for SO_2-emission sums) as apparent emissions over the adjacent North Sea. We interpret the "misplaced" emissions over the North Sea as air mass transport from the North Sea via the emission region in the BENELUX countries to the receptor sites that was not compensated by other low pollution air transport from the North Sea to the stations that had not passed over the Northwestern European emission centers.

Two major atmospheric processes will reduce the concentrations of emitted or in situ formed aerosol particles: dilution through mixing with cleaner air masses and wet scavenging through in-cloud and sub-cloud processes. As a tracer of the first of these two processes Fig. 4a gives the long-term average geographical distribution of trajectory derived wind speed over the study area. Highest average wind speeds and ensuing atmospheric mixing is seen over the major emission centers of Northwestern Germany, the BENELUX countries and adjacent seas whereas lowest wind speeds are seen over Northern Germany and the Southeastern neighbor countries. The long-term average geographical distribution of precipitation as taken by HYSPLIT from the GDAS meteorological fields in Fig. 4b corroborates the results about atmospheric cleaning processes indicated in Fig. 4a. The small absolute numbers in Fig. 4b are due to the episodic nature of precipitation: most of the time it does not rain or snow. The blue crescent reaching from the North Sea through the BENELUX countries, Eastern France, Switzerland and the alpine region exhibits maximum precipitation values while Southern and Eastern Germany with the adjoining countries to the East and Southeast show minimum precipitation values. Thus, in the long term we expect much of the high Western European emissions to be scavenged to a substantially by wet processes. In addition, air masses arriving

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Gelöscht: PM_{10}-emissions are largely concentrated around major conurbations and highly populated and industrialized regions such as the German Ruhr area, and the BENELUX countries whereas highest PM_{10}-concentrations were measured to some extent in air masses from the East and much more so in air masses from the Southeast.

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from Western and Northwestern directions at the stations usually cross the Western European emission centers with much lower pollution burdens than air masses coming from the polluted countries of Southeastern Europe arriving at the corresponding map borders (cf. Fig. PM_{10} — 36th maximum daily average value in µg m^{-3}, 2005 in EEA, 2009).

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5.3. Immission trends for air from specific source regions

As mentioned in the introduction, the pollutant emissions reported by the European and national Environment Agencies represent a synthesis of known pollutant sources combined with assumed emission factors. These emissions are typically used as input for air quality modelling and subsequent assessment, as well as for trend analyses. However, it remains unclear to what extent these reported emissions are realistic, and whether their trends represent the trend in true emissions. Here, we attempt to assess spatially-resolved trends in real particulate emissions by an analysis of measured concentrations (immissions) in air masses travelling over source-specific regions.

To test our method, we selected two pronounced source regions in Europe, located within 1000 km distance from our observation sites. These regions were defined by emission hotspot regions that can be seen in the EDGAR emission maps in Fig. 3a-b and comprise: Region A (Be-NL-NRW; comprising most of Belgium, southern parts of the Netherlands, and much of the German state North Rhine-Westphalia) and Region B (CZ-PL-SK; comprising the central parts of the Czech Republic, southern parts of Poland, and adjacent areas of Slovakia.) According to the European Environment Agency (EEA) these are regions, where reported particulate emissions have developed differently during the past 10 years. Our goal is to verify this through an analysis of real atmospheric observations over this period.

Temporal trends were computed using the customized Sen–Theil trend estimator (Sen, 1968; Theil, 1992). The Sen–Theil estimator is the median of many slopes calculated in a continuous or non-continuous time series, with its robustness against outliers being one of its main assets. For the detailed description of this trend estimator we refer to Sun et al. (2020), Section 2.3.1. Here we computed the Sen–Theil estimator for hourly observation data at stations ME, NG, and WA. Subsets of back trajectories were selected that spent at least 1, 3 , 6, or 12 hours over the source regions A and B. Depending on that criterion, different sub-sets were analyzed. The difference in median eBC mass concentration between air masses arriving from source region A and B is obvious, as could already be determined in the corresponding immission maps (Fig. 2c-d). As we learned from Sect. 5.2 these immission maps are strongly influenced by the different meteorological conditions governing atmospheric dispersion in different wind direction, so that these values allow no direct conclusion on the strength of emission sources located upwind.

We analyzed the temporal trends in eBC over the period 2009-2018 for the subsets belonging to Regions A and B – assuming that these systematic differences in meteorological conditions should even out over such long observation periods. Table 2 shows that the Sen–Theil slope estimator for Region A is between -7.6 % and -5.1 % for the three observation sites and the requirement of a back trajectory to have spent at least 6 hours over Region A. For region B, the corresponding Sen–Theil slope estimators are between -4.0 % and -2.7 % for the observation sites. As we can read from these results, the annual decrease in eBC is more pronounced for air masses that have travelled over Region A.

Between 2009 and 2017 for the EU member states of Belgium, the Netherlands, Germany, the Czech Republic, Poland, and Slovakia the annual rates of decrease in reported emissions were between -4.9 and -6.1 % for the first three countries, and between +0.5 and -2.8% for the

latter three (https://www.eea.europa.eu/data-and-maps/dashboards/air-pollutant-emissions-data-viewer-2). As compiled in Table 2 these reported trends are largely consistent with the rates of changed derived from our eBC immission trends. Although we need to keep in mind that the six nation states only partially contribute to our regions A and B, it seems valid to conclude that BC emissions in region A indeed decreased more rapidly in the past decade compared to region B. Our approach seems able to differentiate between concentrations trends in air masses that have passes over rather different source regions. This might represent a step towards the assessment of changes in real-world emissions allocated in specific source regions over multi-annual periods.

5.4. Comparison of immission and emission trends

Besides the map comparison a second approach was used to connect emission data with the measured aerosol time series. Along each of the hourly back trajectories the emissions according to the EDGAR database were summed up. Then monthly medians of the emission sums and the measured parameters were formed. The EDGAR database reports annual average emissions. PM_{10}, black carbon and other combustion related air pollutants show substantial annual variations with high winter and low summer values at non-urban sites (e.g., Heintzenberg and Bussemer, 2000). In emission modeling the temporal variation of annually reported emissions is considered by disaggregating the annual values with monthly, weekly and daily factors (Matthias et al., 2018). For the time-resolved comparison of PM_{10} and BC-emissions with PM_{10} and eBC-concentrations at the GUAN-sites monthly medians of PM_{10} and eBC-values at the stations were formed and plotted in Fig. 5. We expected both, seasonal variations and a long-term trend in the emissions. For M hours per month of measured components at the four stations the annual average EDGAR-emissions E_{PM10}, E_{BC}, E_{SO2}, and E_{NOx} were summed up along the 121 trajectory steps leading to the stations. Then monthly

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Gelöscht: $PM10$, BC, $SO2$, and NOx-emissions were summed up along the hourly back-trajectories to the stations and monthly medians of these sums

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medians $\tilde{E}_{i=1,4}$ were formed according to Eq. 1 (exemplified for BC). Medians were chosen to reduce the effect of outliers due to local emission and scavenging events.

$$\tilde{E}_{BC} = Median(\sum_{n=1}^{121} E_{BC})_{m=1,M} \quad \text{Eq. 1}$$

The monthly median emission sums $\tilde{E}_{i=1,4}$ were modified with a monthly (f_m) and an annual factor (g_y) in order to simulate respective median monthly measured concentrations taken over all stations. Thus, for each component 12 monthly and 10 annual trend factors determined the agreement of modified summed emissions and measured concentrations. As objective or utility function χ^2 the sum of squared deviations between annually and monthly modified emission sums and monthly median measured concentrations was formed taken over the 120 months of the present study (exemplified for BC in Eq. 2).

$$\chi^2_{BC} = \sum_{j=1}^{120}\left(f_{m=1,12} \cdot g_{y=1,10} \cdot \tilde{E}_{BC} - eBC\right)^2 \quad \text{Eq. 2}$$

χ^2 was minimized with a Generalized Reduced Gradient (GRG) solver (Lasdon et al., 1978) that optimized the12 monthly and 10 annual factors for each of the four measured components. We used Excel's® implementation of the GRG-solver procedure for the optimization. After optimizing month and trend factors the average relative deviation between emission-simulated and measured monthly median curves is 14%, 21%, 25%, and 18% for PM_{10}, eBC, SO_2, and NO_x, and respectively. The optimized monthly median emission sums for all four parameters are displayed in Fig. 5 together with the measured monthly median concentrations.

A ten-year trend in emissions of PM_{10}, BC, SO_2, and NO_x, and average monthly factors for the respective parameters are the two essential results derived from the optimization approach.

[1] nach oben verschoben: We expected both, seasonal variations and a long-term trend in the emissions.

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The ten-year trends relative to 2009 are collected in Fig. 6. Annual averages of the relative differences between the monthly median measured parameters and the corresponding emission derived parameters were formed and applied to the GUAN-trend values displayed in Fig. 6. The resulting error bars on the trends serve as estimates of the uncertainties of the optimization approach. The general trend in Fig. 6 is downward to minima between 30 and 70% of the 2009 values in 2016/17 after which all parameters exhibit increases, most strongly PM_{10}. SO_2 shows the strongest decrease whereas PM_{10} and NO_x-emissions diminished the least. In 2010/2011 the trend curves of PM_{10} and NO_x in Fig. 6 show a slight increase that can be linked to a recovery of economic activity after the world-wide financial and economic crisis during the period 2007-2009. The increase in PM_{10} is also visible in the trend curves relative to 2005 published by the German Environment Agency (https://www.umweltbundesamt.de/daten/luft/luftschadstoff-emissionen-in-deutschland/emissionen-prioritaerer-luftschadstoffe).

The results of two comparisons of our trends with data reported by the German and European Environment Agencies are added to Fig. 6. In general, the trends reported by the German Environment Agency for all German emissions exhibit weaker reductions than the results of the present study. Only for PM_{10} in 2011 and 1013 the present study yields higher values than GEA. We note that primary PM_{10}-imissions may have substantial contributions from wind erosion of agricultural soils (Panagos et al., 2015) that are not incorporated in present anthropogenic inventories. SO_2 exhibits the strongest trend discrepancies with much stronger reductions in trend of the present study as compared to GEA results. As Germany has been reducing SO_2 emissions systematically since the nineteen eighties one would not expect any further strong trends during the time period of the present study. As other studies have demonstrated before, (e.g., van Pinxteren et al., 2019), the maps in Fig. 1 indicate the possibility of imported pollution, in particular from the Southeast. Consequently, we searched for similar

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trends in emission data reported by EEA for neighboring countries until 2017 directly West, South, and East of Germany, going in the East all the way to Romania. Excel's solver optimized combinations of the EEA-trends for Germany and neighboring countries in order to fit the trends derived in the present study. The solver did not choose German trends for any of the four parameters PM_{10}, BC, SO_2, and NO_x. For PM_{10} a combination of emission trends for the BENELUX countries and France was optimum, albeit without being able to simulate the relative maxima in 2011 and 2013 and the minimum around 2016. For BC the emission trend for the BENELUX countries came closest to the trend of the present study. For SO_2 mostly emissions in Romania with minor contributions from French and BENELUX trends simulated the trends observed over Germany best. NO_x-trends were best simulated by emissions over the Czech and Slovakian countries. Emissions trends over Switzerland, Austria, Hungary and Poland were not utilized by the solver. All simulated trends are displayed as curves EEA in Fig. 6. We do not claim that these simulated trends numerically correspond to imported pollution over Germany. However, the good fit of SO_2-trend with emissions over Romany corroborates our finding of pollution import from Southeastern Europe to Northeastern Germany while the development of BC appears to follow better emission trends over Western neighbor countries than over Germany.

Sun et al., (2020) investigated trends of size resolved number and eBC mass concentrations at 16 observational sites in Germany from 2009 to 2018 including the three GUAN-sites of the present study. Based on monthly median time series they report average decreases for ME, NG, and WA of -5.5%, -6.1, and -3.9%, respectively. The corresponding result for eBC of the present study is -4.6%, albeit with a high variability (cf. Fig. 6) of 20 percent units expressed in terms of a standard deviation.

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Over the polluted continent the particle-number based parameter N_{10-800} is largely secondary in nature, i.e., its concentrations are controlled by atmospheric constituents and processes. Thus, there is no primary emission data base with which a similar trend analysis as with PM_{10}, BC, SO_2, and NO_x could be attempted. Instead we chose the 10-year Grand Averages (GA) averages taken over the whole time period of the present study as references from the deviations of annual averages are discussed. Sun et al. (2020) report very minor trends (between -3.5% and 0.1%) for N_{20-800} at the three GUAN stations of the present study. The 10-year interannual variation of our N_{10-800} in Fig. 7a) bears out why only a minor trend if any can be expected. For the first four years the annual averages are substantially higher than average. Then annual values decrease down to a minimum in the years 2016/17 before they increase again to a level slightly above the 10-year average.

In Figs 7b-d) annual deviations from the respective GAs are displayed that can be connected to the 10-year course of N_{10-800}. Ozone concentrations averaged over the data from the three GUAN stations can be interpreted as an indicator for photochemical activity that also controls NPF. The annual deviations of O_3 in Fig. 7b) follow rather closely those of N_{10-800}. In Figs 7c) and d) annual deviations of ambient temperature and precipitation rates are displayed that have been averaged over the meteorological data along the back trajectories leading to the four stations. For the temperature an averaging period of 120 trajectory hours yielded the highest (negative) correlation with N_{10-800} of r = -0.8. After a dip in 2009 annual average trajectory temperatures to a maximum in 2016 before returning to near average in 2018. For the precipitation rates along the trajectories the highest (negative) correlation with N_{10-800} was found with an averaging period of three days (r=-0.6) before arrival at the stations. The results displayed in Figs 7c) and d) illustrate the complexity of processes and conditions controlling atmospheric particle number concentrations. On one hand, a scavenging effect of precipitation can be used as argument for the high values of N_{10-800} in the years 2010-2013 and the low values

in the years 2014 through 2018. On the other hand, lower annual temperatures during years of relatively high N_{10-800} and higher than GA-temperatures during years of relatively high N_{10-800} are harder to interpret. Possibly the nucleation of condensable vapors is furthered by lower air temperatures upwind of the stations.

An important result of trend analysis are the average monthly factors disaggregating the annual emissions. In general the summer minima of the month factors determined in the present study are broader than the curve given by Matthias et al., (2018) for combustion emissions. The decrease of the month factor of PM_{10}, BC, and NO_x in December and the late winter maxima of PM_{10} and SO_2 are not reflected in the Matthias et al., (2018) results. Interestingly, both PM_{10} and SO_2 show a minor secondary peak in June. As an example of the seasonal variability of eBC within an urban source region we averaged the relative annual variation of eBC-concentrations at the station Leipzig Eisenbahnstraße (plotted as curve L-EBS in Fig. 8) exhibiting a smaller seasonal swing than all other curves. The curve for PM_{10} comes closest to that for L-EBS, probably because of agricultural non-combustion emissions in summer.

In general the downward trends in particulate parameters determined in the present study are similar to temporal trends of particle number and black carbon mass concentrations at 16 observational sites in Germany from 2009 to 2018 (Sun et al., 2020). The long-term emission-decrease of PM_{10} as determined in the present study from 2009 to 2018 is smaller than the corresponding number published by the EEA as average over all 28 EU member-states but similar to the figures published by GEA until 2017 (cf. Table 2). For BC, SO_2, and NO_x the present study yields substantially stronger emission-reductions than both GEA and EEA. These findings are emphasized when considering 2017 as endpoint of the trend calculation (cf. Table 2) at and after which our study shows consistent emission increases of all studied parameters. Comparing the calculated trends with emission trends in neighboring countries as published by

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706 the European Environment Agency supports the explanation that the observed trends are to
707 some extent due to changes in imported air masses. Most strongly this holds for SO_2, the trend
708 of which follows that of Romanian emissions rather well.
709
710 The last issue we take up in this discussion concerns the frequent residual difference between
711 measured and emission-simulated time series. In Fig. 5, e.g., in most winters there are months
712 when optimized BC-emissions remain substantially lower than the measured monthly medians
713 of eBC. Some information can be gleaned from the "Großwetterlagen", (GWL), representing
714 29 classifications of large scale weather types after Hess and Brezowsky for Central Europe,
715 (Gerstengarbe and Werner, 1993), provided by the German Weather service for each day
716 (http://www.dwd.de/DE/leistungen/grosswetterlage/grosswetterlage.html). During the winter
717 months with the strongest difference between measured and simulated time series the
718 probabilities of high-pressure systems over Fennoscandia with south-to-southeasterly flow to
719 the four stations is substantially higher than the respective probabilities averaged over the whole
720 ten-year period of the study. This GWL-information is consistent with the back trajectories
721 during the high pollution winter months coming predominantly from the southeasterly sector
722 of the map. While the classified large-scale weather situation with weak dilution of pollution
723 during the winter months is conducive of high particulate concentrations at the receptor sites it
724 does not explain the discrepancy. In principle our simplistic approach of accumulating
725 emissions along back trajectories may be flawed during certain weather situations. However,
726 an alternative explanation could be that the emissions inventories over Eastern and Southeastern
727 Europe in the EDGAR database are somewhat lower than the real emissions.
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730 **6 Summary and conclusions**

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Ten years of hourly aerosol and gas data at three stations of the German Ultrafine Aerosol Network GUAN and one station of the Saxonian Environment Agency have been combined with hourly back trajectories to the stations and emission inventories. Measured PM_{10}, particle number concentrations between 10 and 800 nm, and equivalent black carbon were extrapolated along the trajectories. This process yielded what we termed immission maps of these aerosol parameters over Germany. They reflect aerosol emissions modified with atmospheric processes along the air mass transport between sources and the four receptor sites at which potential effects of the particulate air pollution would be realized.

The ten-year average immission maps do not simply show the distribution of pollution sources upwind of the receptor sites. The comparison with emission data based on the European EDGAR emission database shows that strong Western European emission centers do not dominate the downwind concentrations because their emissions often are reduced by wet scavenging and dilution processes on the way to the receptor area. Maps of average precipitation and wind as they occurred along the trajectories illustrate these processes. In the receptor region mass related aerosol parameters such as PM_{10}, equivalent black carbon, and to some extent also the particle number concentration instead is rather controlled by emissions from Eastern and Southeastern Europe from which pollution transport often occurs under dryer meteorological conditions in continental high-pressure air masses. This finding corresponds to the air mass results derived for the sub-micrometer particle number size distribution by Birmili et al., (2001), by Engler et al., (2007) for the size distribution of non-volatile particles, by Ma et al., (2014) for optical particle properties all evaluated at the station Melpitz, and by van Pinxteren et al., (2019) for transboundary transport of PM_{10} to ten stations in Eastern Germany from neighboring countries. Newly formed particles on the other hand are found in air masses from a broad belt reaching from Burgundy to the Western Czech Republic and Southern Poland

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765 a region with high photochemical activity in summer that is affected by emissions in Northern
766 Italy.
767
768 Annual EDGAR emissions for 2009 of PM_{10}, BC, SO_2, and NO_x, were accumulated along
769 each trajectory and compared the calculated emission sums with the corresponding measured
770 time series on a monthly basis. With a generalized reduced gradient solver the agreement of
771 each pair of monthly time series e.g., measured eBC and BC-emissions was optimized by letting
772 the solver determine both monthly emission factors disaggregating the annual EDGAR
773 emission fields and adjusting the emissions with annual factors modifying the 2009-fields.
774 Relative to 2009 the annual averages of the analyzed air pollutants were lower in 2018 by values
775 between 6% for PM_{10} and 60% for SO_2. In general, the ten-year reductions determined of the
776 present study were stronger than those reported by the German and the European Environmental
777 Agencies. $N_{10\text{-}800}$ exhibited substantial interannual variability but no net decrease over the ten
778 studied years.
779
780 The validity of the present approach of connecting immission and emission of particulate
781 pollution was tested by calculating temporal changes of eBC for subsets of back trajectories
782 passing over two separate prominent emission regions, region A to the Northwest and B to the
783 Southeast of the measuring stations. Consistent with reported emission data the calculated
784 immission decreases over region A are significantly stronger than over region B.
785
786 Compared to published emission monthly factors by Matthias et al., (2018) the present
787 approach yielded broader summer minima that were partly displaced from the midsummer
788 positions given by Matthias et al., (2018). As an aside we note that during the winter months
789 with extremely high particulate pollution the emissions accumulated along back trajectories

Gelöscht: which we explain with gaseous particle precursors being transported with little wet scavenging from this region

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often are substantially lower than the measured concentrations which raises the question of the validity of the emission figures in Eastern and Southeastern European source regions.

There are clear limits in the methodology of the present study. Air mass trajectories have inherent uncertainties increasing with their distance travelled (Stohl, 1998). Meteorological processes affecting the aerosol during air mass transport are only considered rather coarsely whereas aerosol dynamics are not considered at all. Possible future improvements concern ensemble trajectories with higher resolution, better meteorological information along the trajectories e.g., radar-derived precipitation as used in Heintzenberg et al., (2018), more comprehensive emission inventories with higher spatiotemporal resolution and higher numbers of analyzed stations.

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Gelöscht: support given

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Table 1: Characteristics of the four stations of the present study, see text for instrumental details. The number of validated data hou given for each component

Station	Acronym	Latitude	Longitude	MPSS[1]	eBC[2]	PM10 continous[3,4]	PM10 discontinous[5]	NO_x[6]	SO_2[7]	O_3[8]
Collmberg	CO	51.3	13			85054		88838		88792
Melpitz	ME	51.5	12.9	81561	88196		88822	86260	85541	84421
Neuglobsow	NG	53.1	13	57962	77540	71202		83718	87778	87943
Waldhof	WA	52.8	10.8	84276	80725	88321		85503	82386	87373

[1]MPSS - scanning mobility particle size spectrometer TROPOS (10 – 800 nm); [2]MAAP - Multi-angle absorption photometer 5012 Thermo Fischer Scientific; [3]TEOM-FDM - Tapered element oscillating microbalance fitted with a filter dynamics measuring system 1405 Thermo Fischer Scientific; [4]SCHARP - Synchronized Hybrid Ambient Real-time Particulate Monitor 5030 Thermo Fischer Scientific; [5]HVS – High Volume Sampler DIGITEL DH-80; [6]TLA-NOx –Trace Level NOx Analyzer 42i-TL Thermo Fischer Scientific; [7]TLA-SO2 - Trace Level SO_2 Analyzer 43i-TLE Thermo Fischer Scientific; [8]

Table 2 Median concentrations of eBC concentrations (µgm^{-3}) and temporal trends (2009-2018) of eBC in terms of Sen-Theil slope (% per year) as determined for air masses passing over Regions A and B as analyzed at the stations Melpitz (ME), Neuglobsow (NG), and Waldhof (WA). For comparison the national annual decreases in BC emissions 2009-2017 in % according to the European Environmental Agency are added.

	DELTA T*	Median eBC in										Decrease in national BC emissions in % per year		
		No. of back trajectories			µm/m³			Sen-Theil slope in % per year						
	in h	**ME**	**NG**	**WA**	**ME**	**NG**	**WA**	**ME**	**NG**	**WA**	**3 Stations****	Belgium	Netherlands	Germany
	1	21941	17514	27218	0.38	0.40	0.41	-6.40	-6.80	-4.80	-5.85	-6.1%	-6.1%	-4.9%
Region A	3	18605	14268	22132	0.38	0.40	0.41	-6.40	-6.90	-4.80	-5.89			
B-NL-NRW	6	14802	10086	15936	0.39	0.40	0.42	-6.40	-7.60	-5.10	-6.19			
	12	6817	3746	6131	0.40	0.50	0.50	-7.10	-7.90	-5.30	-6.62			
												Czech Rep.	Poland	Slovakia
	1	11096	5264	4191	1.10	1.19	1.13	-3.60	-3.40	-1.70	-3.16	-2.8%	0.5%	-2.3%
Region B	3	9601	4339	3541	1.08	1.18	1.12	-3.40	-3.40	-2.10	-3.14			
CZ-PL-SK	6	7000	3062	2570	1.05	1.09	1.11	-4.00	-2.90	-2.70	-3.47			
	12	3628	1410	1277	1.00	1.00	1.00	-3.70	-3.00	-2.70	-3.34			
ALL		85846	75190	78356	0.45	0.36	0.36	-5.90	-5.60	-4.00	-5.18			
Sun (2020)								-4.40	-7.80	-3.20				

* Minimum time spent over the specified source region, **Weighted mean, according to the available number of back trajectories

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Table 3 Percental decreases in the anthropogenic emissions of PM_{10}, BC, SO_2, and NO_x relative to 2009 as reported by the European Environment Agency (EEA, https://www.eea.europa.eu/data-and-maps/dashboards/air-pollutant-emissions-data-viewer-2), the German Environment Agency (GEA), and calculated in the present study. The EEA and GEA only report data until 2017, (*=BC until 2016).

Component	EEA 2009-2017	GEA 2009-2017	GUAN emissions 2009-2017	GUAN emissions 2009-2018
PM_{10}	12%	4.2%	16%	6%
BC*	29%	35%*	63%	44%
SO_2	33%	20%	68%	59%
NO_x	20%	11%	43%	30%

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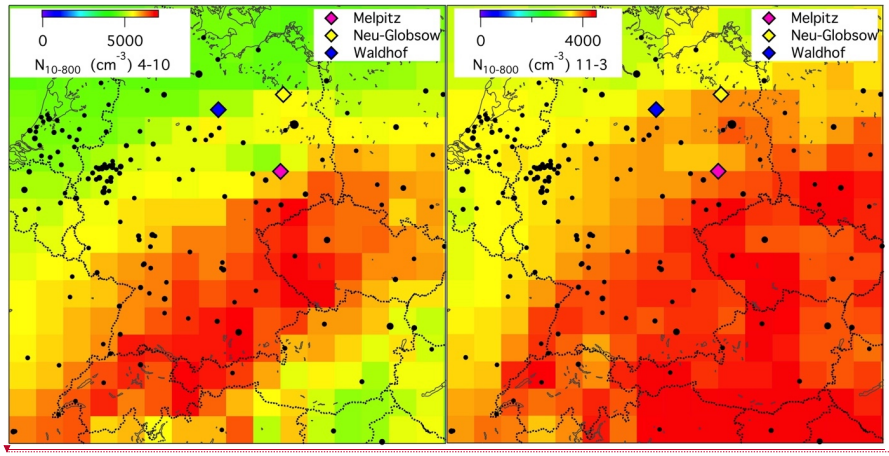
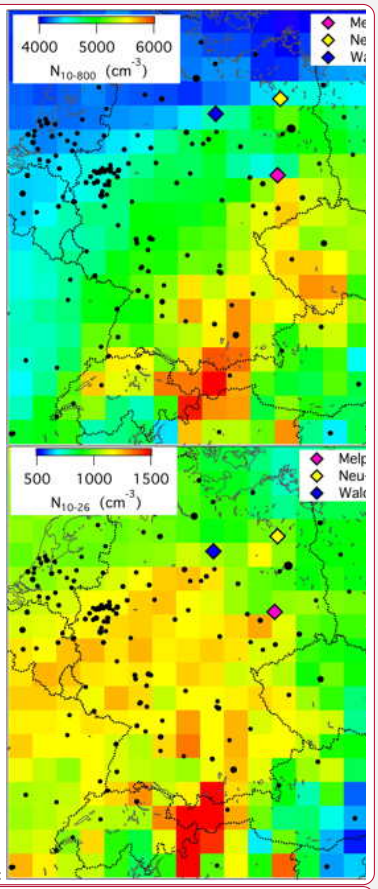


Fig. 1 Maps of particle number concentration N_{10-800} (cm^{-3}) extrapolated under 1000 m height along five day back trajectories from hourly data at the four stations from 2009 to 2018; left: months April through October; right: months November through March. The GUAN-stations are marked with colored diamonds. The Collmberg station lies 30 km Southeast of station Melpitz. Here and in the following maps the black dots represent cities larger than 100000 inhabitants with the size of the dots being proportional to the number of inhabitants.



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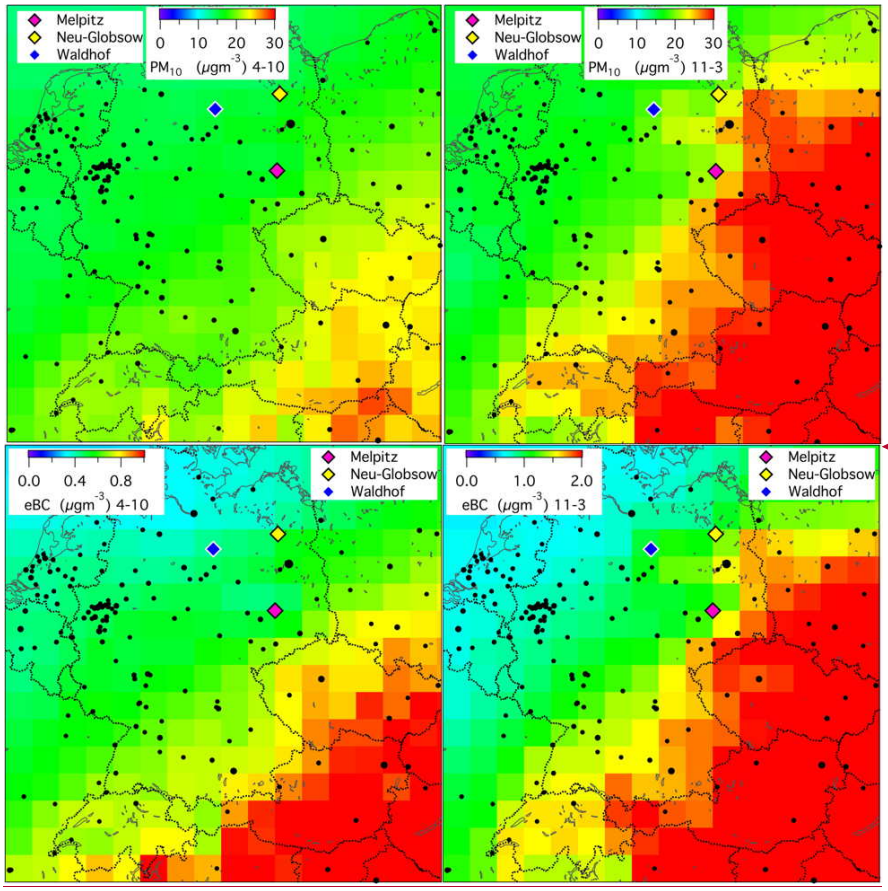

Fig. 2 As Fig. 1 but for particle mass concentrations (top, PM_{10}, µgcm^{-3}), and black carbon concentrations (bottom, eBC, µgm^{-3}),

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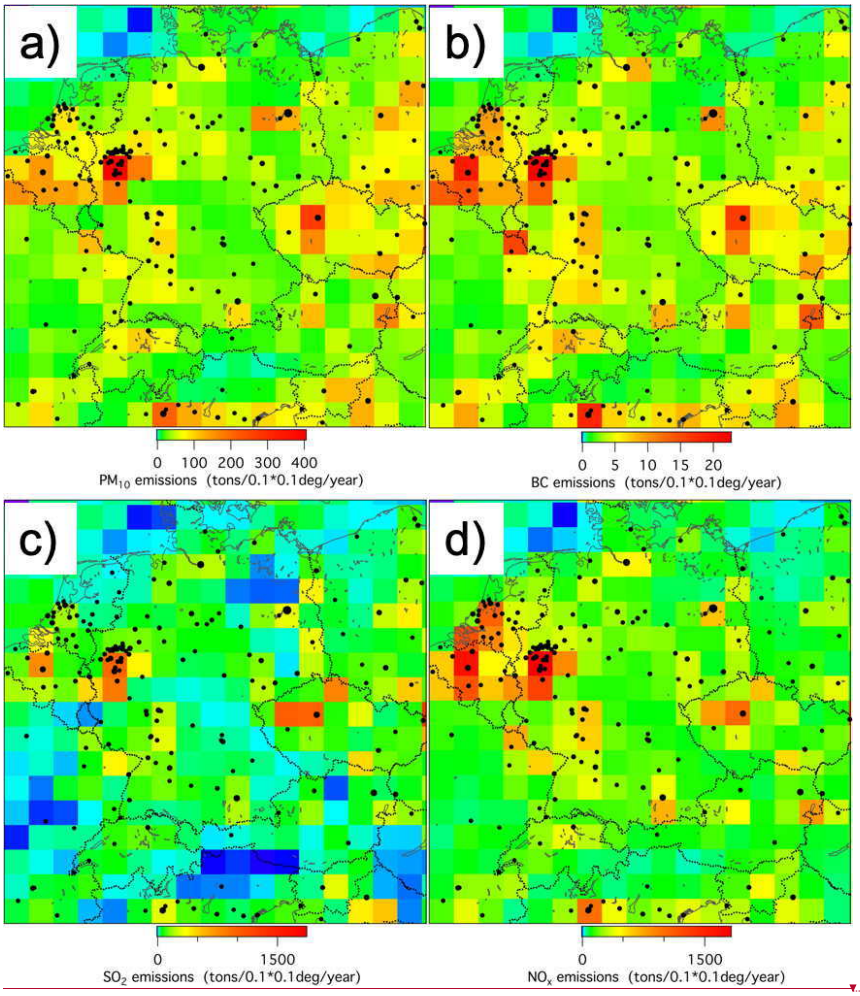

Fig. 3 As Fig. 1 but a) for PM_{10} emissions (tons/0.1*0.1deg./year), b) for BC emissions, c) for SO_2 emissions, and d) for NO_x emissions (tons/0.1*0.1deg./year) according to the EDGAR emission database (https://data.europa.eu/doi/10.2904/JRC_DATASET_EDGAR) for 2009 averaged over the geogrid of the present study.

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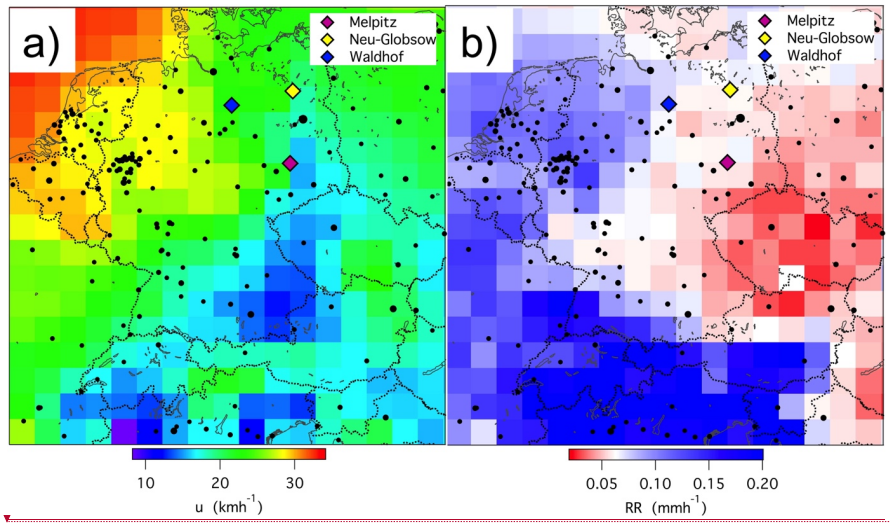

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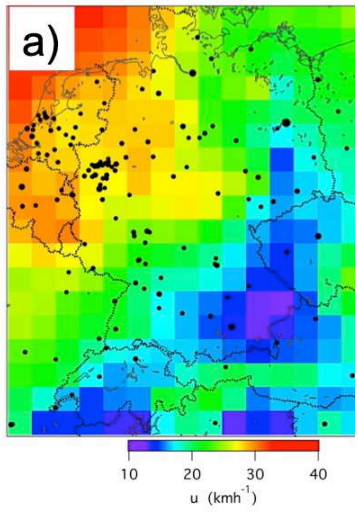

Fig. 4 a) Map of horizontal wind speed (u, kmh^{-1}) as reported by HYSPLIT along hourly five-day back trajectories to the four stations marked in the graph averaged over the time period 2009 to 2018; b) as a) but for precipitation (RR, mmh^{-1}).

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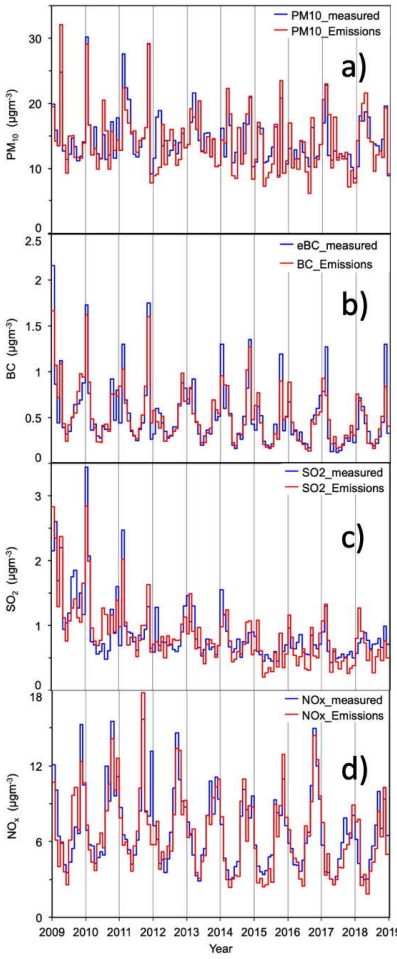

Fig. 5 a) Monthly medians of PM_{10}-concentrations at the four stations of the present study (blue), and monthly medians of optimized sums of PM_{10}-emissions along back trajectories leading to the stations (red). b) as a) but for measured eBC-concentrations and BC-emissions along back trajectories. c) as a) but for measured SO_2-concentrations and SO_2-emissions along back trajectories. d) as a) but for measured NO_x-concentrations and NO_x -emissions along back trajectories.

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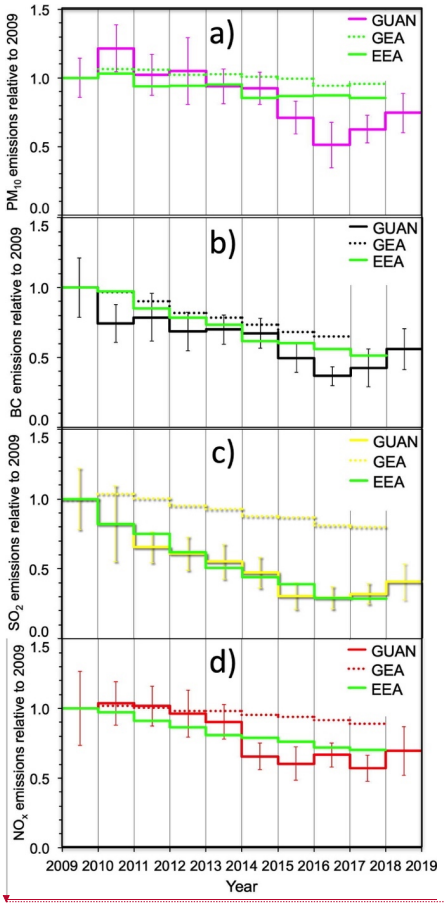
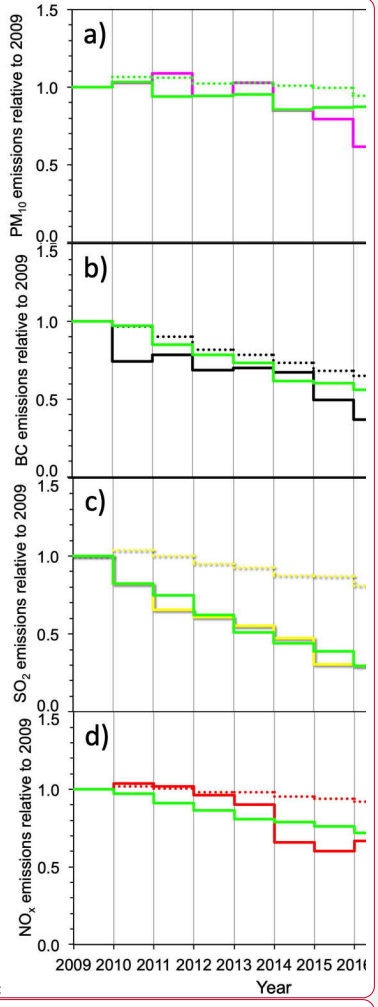


Fig. 6 GUAN: Trends in the emissions of a) PM_{10}, b) BC, c) SO_2, and d) NO_x, relative to 2009 as calculated by optimizing the agreement between 2009-EDGAR-emissions and concentrations measured at the four stations of the present study. The error bars represent annual average relative deviations between measured and simulated data. GEA: Trends as reported for Germany by the German Environment Agency. EEA: Trends as optimized from combinations of trends over Germany and neighboring countries, (see text for details).



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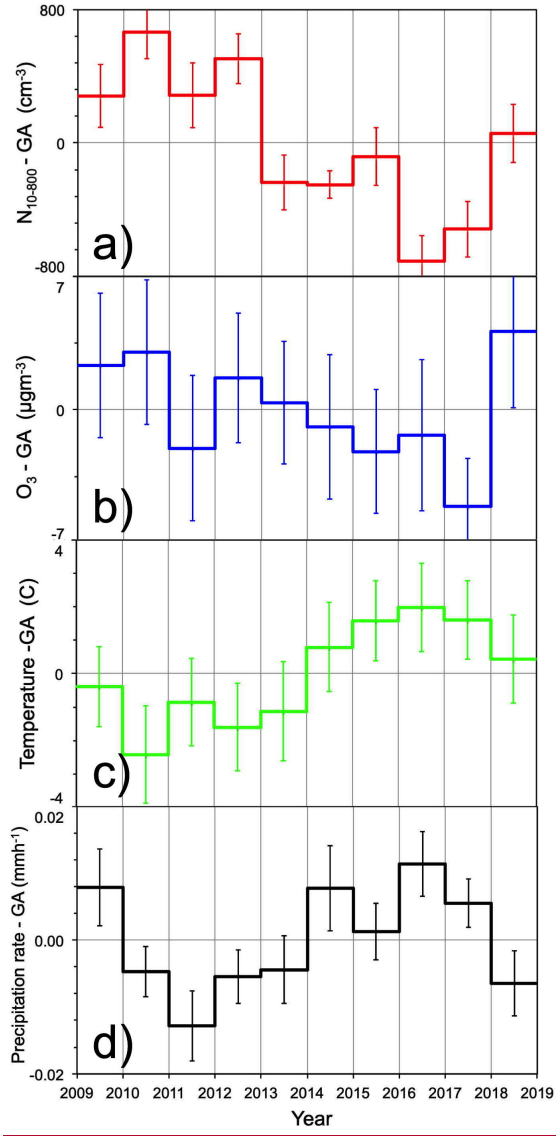

Fig. 7 Trends in annual average deviations a) ΔN_{10-800}, b) ΔO_3, c) temperature ΔT along the trajectories five days back in time, and d) precipitation rate ΔRR along the trajectories three days back in time. The deviations are taken relative to the respective 10-year

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Formatiert: Schriftart: Symbol

Grand Average (GA). The error bars represent the standard deviations of the annual averages.

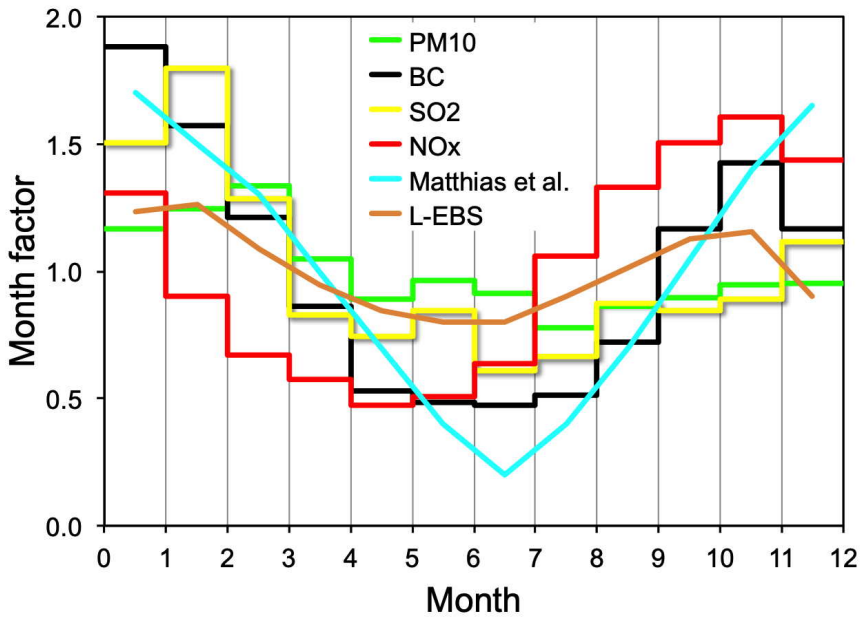
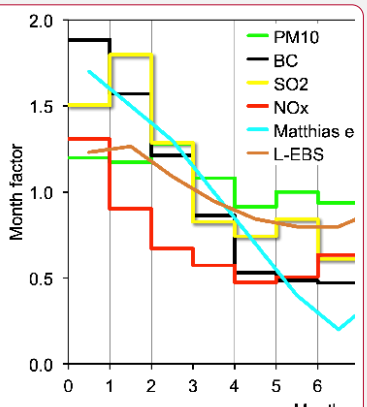


Fig. 8 Month factors for the emissions of PM_{10}, BC, SO_2, and NO_x as determined by optimizing the agreement between EDGAR-emissions and concentrations measured at the four stations of the present study. For comparison the month factors of Matthias et al., (2018) for combustion emissions are plotted and the relative annual variation of eBC concentrations measured at the station Leipzig-Eisenbahnstraße (L-EBS) averaged over the time period of the present study.

Gelöscht:



Gelöscht: 6

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Seite 40: [2] Formatiert	Nemo	01.04.20 11:58:00

Rechts: 2 cm, Unten: 2.5 cm, Breite: 29.66 cm, Höhe: 20.99 cm

Seite 40: [3] Formatierte Tabelle	Nemo	30.04.20 11:06:00

Formatierte Tabelle

Seite 40: [4] Gelöscht	Nemo	01.04.20 11:49:00

Seite 40: [4] Gelöscht	Nemo	01.04.20 11:49:00

Seite 40: [5] Formatiert	Nemo	02.04.20 15:32:00

Schriftart: 12 Pt.

Seite 40: [6] Formatiert	Nemo	02.04.20 15:32:00

Schriftart: 12 Pt.

Seite 40: [7] Formatiert	Nemo	30.04.20 11:06:00

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Seite 40: [7] Formatiert	Nemo	30.04.20 11:06:00

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Seite 40: [8] Formatiert	Nemo	02.04.20 15:32:00

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Seite 40: [8] Formatiert	Nemo	02.04.20 15:32:00

Schriftart: 12 Pt.

Seite 40: [9] Formatiert	Nemo	02.04.20 15:32:00

Schriftart: 10 Pt.

Seite 40: [10] Formatiert	Nemo	02.04.20 15:32:00

Schriftart: 10 Pt.

Seite 40: [11] Formatiert	Nemo	30.04.20 11:07:00

Hochgestellt

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Hochgestellt