Dear editor:

We would like to thank the two referees for their helpful comments and suggestions, which have been fully taken into account upon manuscript revision. A point-by-point response to all the comments and a revised manuscript were uploaded.

Sincerely yours,

Nan Ma and co-authors

Response to comments of referee #1

General comments:

The authors reported historical trends of particle number and black carbon concentrations. The article is well-written. I recommend to publish in ACP. A major comment is given here. The authors try to link the pollutants historical trends with mitigation policies. However, only few pollution control regulations are introduced in the introduction section. These regulations should be also mentioned to explain the changes in pollutant concentrations. Thus, it would be much better for understanding the long-term effects of emission mitigation policies. Some implications on the benefit of decreasing PN and BC, for examples, health effects and visibility, should be added at the end of article to enhance the scientific and policy significance.

Response:

Many thanks for the comments and suggestions.

Following your suggestion of "regulations should be also mentioned to explain the changes in pollutant concentrations" and the general comment 1 of the referee #2, we have rewritten most part of the result section of the manuscript, trying to find more connections between the observed trends and emission variations. A new section "**3.2 Emission change in Germany**" has been added in the manuscript, in which the overall trends of eBC mass concentration and PNCs are compared with the emission data reported by the Federal Environment Agency. Section "**3.3 Diurnal variation of trends**" and "**3.4 Seasonal variation of trends**" have been rewritten and we tried to find the connections between the diurnal and seasonal trends of observed parameters and the sources which have also distinct diurnal or seasonal variations. As a special case study, a new section "**3.5 Evaluation of low emission zones**" has been added in the manuscript to figure out if such a long-term observation network can reflect the effect of a specific emission mitigation policy.

Following your suggestion "Some implications on the benefit of decreasing PN and BC, for examples, health effects and visibility, should be added at the end of article to enhance the scientific and policy significance", a short discussion has been added at the end of Sect. 3.2:

"Based on the above results, we believe that the observed trends of PNCs and eBC mass concentration are mainly due to the reduction in emissions. The annual changes of meteorological conditions might have an impact on PNCs, but are not likely to be the decisive impact factor. Detailed discussion on the possible influence of meteorological conditions will be discussed in Sect. 4. The decreased pollutant concentrations are highly associated with the reduced risk of human health. Pope et al. (2009) demonstrated that a decrease of 10 μg m^{-3} in the $PM_{2.5}$ mass concentration is related with an increase of life expectancy of 0.61 ± 0.20 year in 211 countries. The improved health effects because of decreased UFP and BC would be even greater compared with that of $PM_{2.5}$ mass concentration. As of 2018, 97 % of cities in low- and mid-income countries do not meet the World Health Organization (WHO) air quality guidelines (WHO, 2018). Our result demonstrates that the implementation of proper emission mitigation policies can largely reduce the BC mass concentration and PNC, thus may effectively reduce the health risk in polluted regions."

__**Response to comments of referee #2**__

General comments:

(1) The paper presents a valuable dataset and analyses long-term trends of PNC and BC across Germany. The topic is within the scope of ACP, but in my opinion the scientific question does not have to be only how is it changing but why is that as well. Some of the presented conclusions could be better supported by data. For example, if the driver of the PNC and BC decrease is expected to be an emission decrease, could this be compared to any reported emission data? Or any mitigation strategies results? Comparison of the trends with such data would be an added value to the manuscript.

Response:

Thanks for your suggestions. We have rewritten most part of the result section in the manuscript. A new section "**3.2 Emission change in Germany**" has been added in the manuscript, in which the overall trends of eBC mass concentration and PNCs are compared with the emission data reported by the Federal Environment Agency (Fig. 3 in the manuscript). We found that total emission of BC in Germany deceases about −3.4 % per year during 2009−2017 and highly agrees with the trend of observed eBC mass concentration, suggesting that emission reduction is very likely to be the dominant factor for the decrease in eBC mass concentration over Germany. The total emission of $PM_{2.5}$ and precursors show decreasing trends as well. However, the decreases in PNCs are stronger. This discrepancy is thought to be caused by the highly complex and nonlinear processes of secondary aerosol formation. Based on the comparisons, we believe that the observed trends of eBC mass concentration and PNCs are mainly due to the emission reduction.

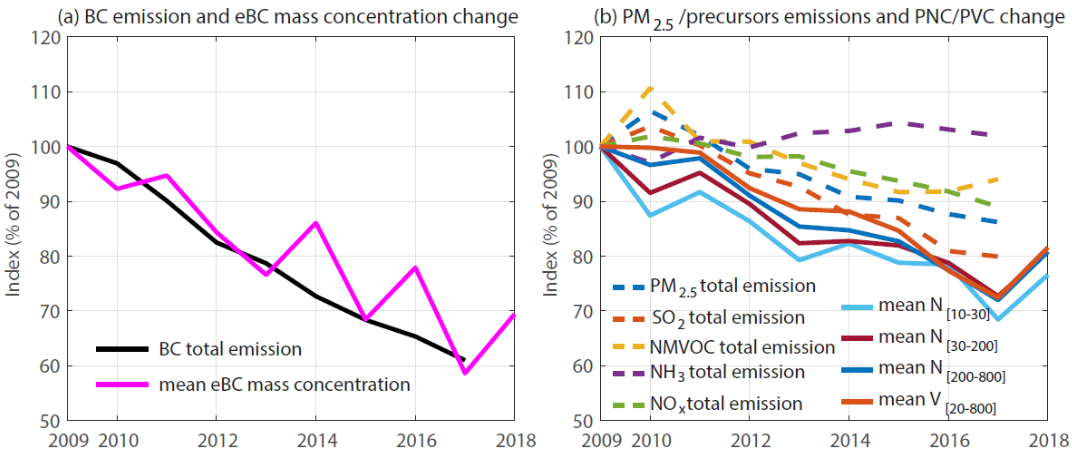

Figure 3: Comparison of the long-term changes in measured parameters and total emissions in Germany.

The emission intensities of some sources have distinct diurnal or seasonal variations, such as that of traffic and residential activities. Section "**3.3 Diurnal variation of trends**" and "**3.4 Seasonal variation of trends**" have been rewritten and we tried to find out the connections between the diurnal and seasonal trends of observed parameters and those sources.

As a special case study, a new section "**3.5 Evaluation of low emission zones**" has been added in the manuscript. The reason for adding this section is, the observed decrease in trends is usually a combined result of the various emission mitigation policies. A question raised is that can such long-term observation network reflect the effect of a specific emission mitigation policy. We select one special mitigation policy low emission zone (LEZ) and trying to figure out its effectiveness based on our dataset. We find gradual decreases in eBC mass concentration and $N_{[30-200]}$ after the implementation of LEZ. But even with the seasonal variation subtracted from the time series, the amplitude of variation of eBC mass concentration and $N_{[30-200]}$ is still very large mostly due to variations in meteorological conditions (Fig. 6 in the manuscript). However, very clear responses are found in the increment of the aerosol concentration (the difference of the concentrations between the traffic and the background sites) (Fig. 7 in the manuscript), suggesting that with a multiple-site network, the effect of emission control policy could be better detected from the increments between near-source and background sites.

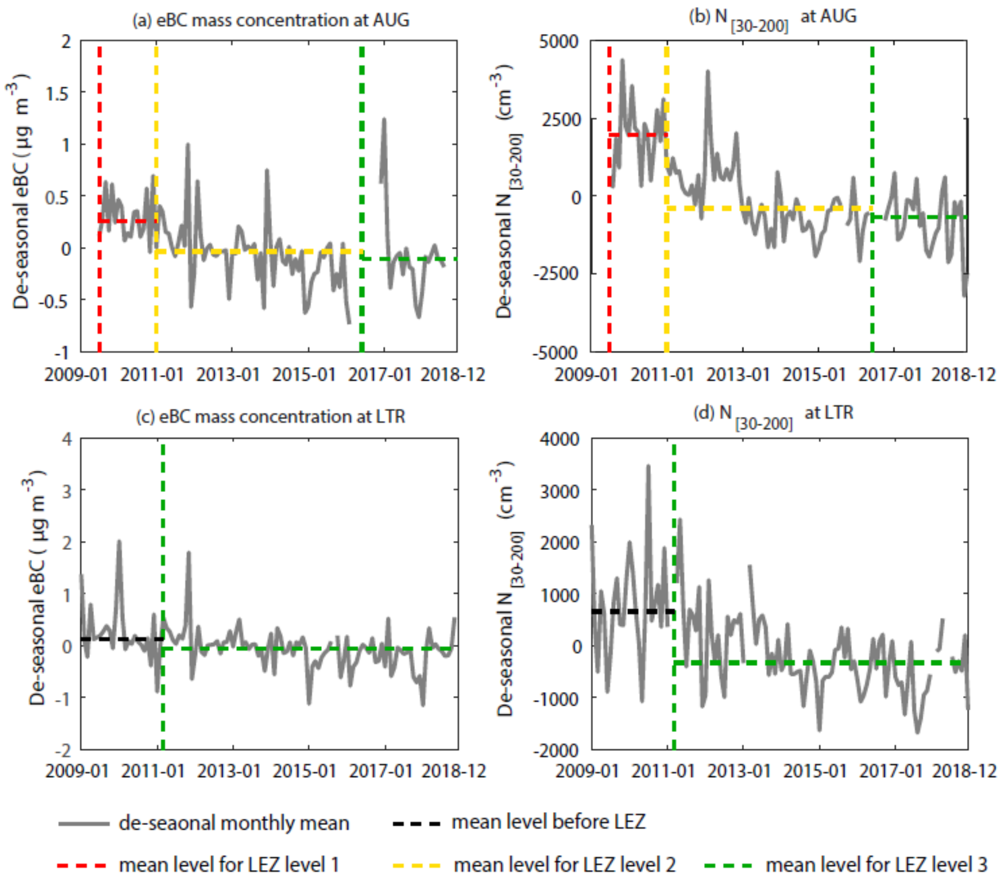

Figure 6: De-seasonalised monthly time series of eBC mass concentration and $N_{[30\text{-}200]}$ at the two urban background sites AUG and LTR. The vertical dashed lines refer to the start dates of LEZ of different stages in the city of Augsburg and Leipzig. The horizontal dashed lines refer to the mean concentration levels of measured parameters during the corresponding time period.

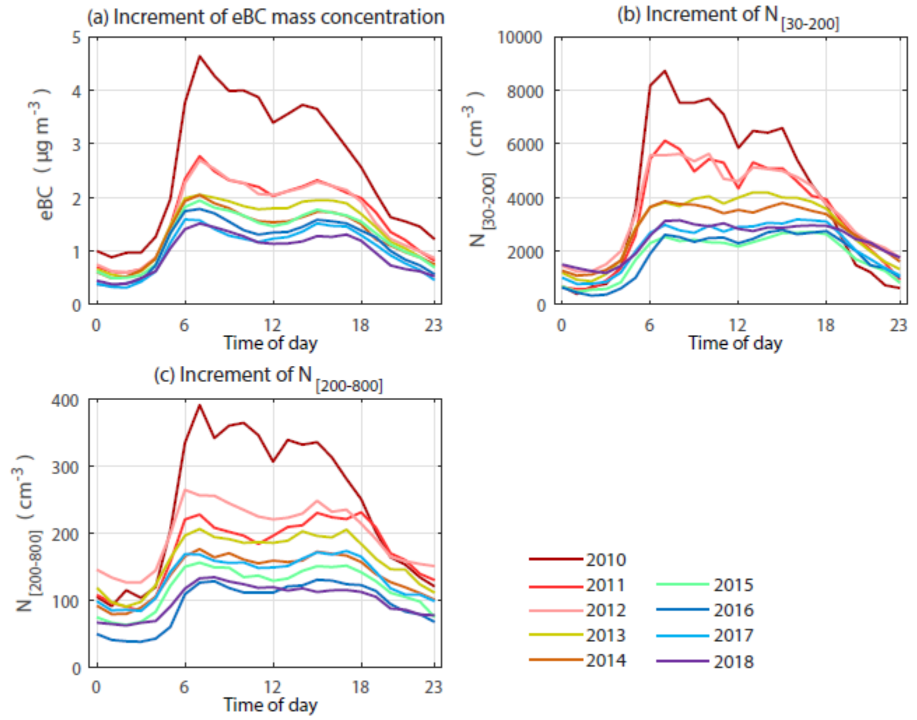

Figure 7: Average diurnal cycles of the increment (defined as the difference between LMI and LTR) in eBC mass concentrations, $N_{[30\text{-}200]}$ and $N_{[200\text{-}800]}$.

(2) Quite some information on BC has been already published in a more detailed paper by Kutzner et al, with more stations and longer dataset. It would be good to include an explanation on what this manuscript brings in addition to the already published results?

Response:

Thanks for the suggestion. There are several differences between Kutzner et al. (2018) and our study.

(a) Data from 12 stations in 3 states were used in Kutzner et al. (2018) for the trend analysis. In our study, 16 stations distributed in 8 states (Fig. 1 in the manuscript) were used and therefore can better represent the variation of BC concentration across Germany.

(b) Most of the stations (11 out of 12) are located in urban area and largely influence by traffic. Thus, only the mitigation of traffic emission was considered in explaining the trend of BC in Kutzner et al. (2018). In our study, stations in various environments (3 roadside, 5 urban background, 3 regional background, and 3 mountain area) are used, and the changes of different sources (traffic, domestic heating, other fuel combustions, industry, etc.) were considered.

(c) Only general trend of BC is reported in Kutzner et al. (2018). In our study, the diurnal and seasonal variations in the long-term trend of BC were investigated and connected to the change of specific emissions.

(d) A comprehensive evaluation of the influence of the inter-annual variations of meteorological conditions (e.g., air masses, precipitation and temperature) is presented in our study.

Following your suggestions, the following sentences have been added at the end of Sect. 1 in the manuscript:

"Kutzner et al. (2018) evaluated the trend of BC over Germany based on measurement at traffic, urban background, and rural sites for the period of 2005–2014, and concluded that the observed decreasing trends in BC are likely owing largely to mitigation measures in the traffic sector. However, there is still a lack of a thorough investigation of the connections between the long-term trends of PNCs/BC and the change of different anthropogenic emissions. A better understanding of the influence of the inter-annual variation of meteorological conditions on the observed trends is also needed.

Based on a unique dataset from the German Ultrafine Aerosol Network (GUAN), this study investigates the long-term variation in the regional PNC and BC mass concentration, to understand the effectiveness of the emission mitigation policies in reducing the PNC and BC in Germany. The study was conducted for the period of 2009–2018 with data from 16 observational sites representing different types of environment (roadside, urban background, regional background, low mountain, and high Alpine). The overall, diurnal, and seasonal trends of PNCs and BC are evaluated and the role of potential decisive factors, including not only emission mitigation policies, but also other potential drivers (i.e. inter-annual change in meteorological conditions and long-range transport patterns) are discussed."

Moreover, a comparison between our results and other studies is given in Sect. 3.1:

"The trends of the PNCs and eBC mass concentration in this study are consistent with results from other studies conducted in Europe. Table 4 compares the long-term trends of aerosol concentrations between the present and other studies. Since 2001, the s.s. decrease in BC, PNCs, and $PM_{2.5}$ have been detected for most of the evaluated sites in Table 4. The implementation of emission mitigation polices have been thought to be the dominant impact factors in these studies. Especially, there is one similar study that evaluated the trend of BC mass concentration in Germany for the time period 2005−2014 (Kutzner et al., 2018), in which decreased BC mass concentration was detected in 12 sites. Comparing the two studies, the absolute decreasing trend of BC mass for 2005-2014 is stronger than our results for 2009−2018, which might stem from the difference between the effects of emission mitigation policies in the two study periods."

Table 4. Comparison of long-term trend studies of BC, PNC, and PM in Europe.

Study	Time period	Region	Parameters	Annual slope (numbers in brackets are the absolute slope, in µg m^{-3} $year^{-1}$)
This study	2009−2018	Germany	BC	Traffic (3 sites): −11.3 %~−5.0 %, (−0.19~−0.08); UB (5 sites): −8.1 %~−2.3 % (−0.08~−0.03); RB to high Alpine (6 sites): −7.8 %~−1.7 % (−0.03~0.00)
			$N_{[20-800]}$	Traffic (3 sites): −7.3 %~−2.9 %; UB (7 sites): −6.3 %~−2.6 %; RB to high Alpine (6 sites): −4.2 %~−0.2 %
Kutzner et al., 2018	2005−2014	Germany	BC	Traffic (7 sites): (−0.31, −0.15); UB (4 sites): (−0.1, −0.02); Rural (1 site): 0.00
Asmi et al., 2013	2001−2010	Europe	$N_{[20-800]}$	Rural to remote (4 sites): −4.6 %~1.6 %
Collaud Coen et al., 2013	2001−2010	Europe	Absorption coef.	Rural to remote (4 sites): −1.6 %~0.0 %
Bigi and Ghermandi, 2016	2005−2014	Italy, Po valley	$PM_{2.5}$	Traffic (2 sites): −6.4 %~−4.6 %; UB (17 sites): −8.1 %~ −0.4 %; RB (4 sites): −4.9 %~0.0 %
Singh et al., 2018	2009−2016	United Kingdom	BC	Traffic (1 site): −8.0 %; UB (2 sites): −5.0 %~−4.7 %; Rural (1 site): −7.7 %

(3) The methods and data quality are mostly appropriate with some exceptions. The number of evaluated stations is changing during the text. Why there are stations with no or non-analysed data? If LWE is not evaluated, why is it included in the text? If LAN and Raunheim stations are used for some analyses, why have not these been used from the beginning? At L278, 5 parameters at 16 sites makes 80 trends, why only 77 of them was evaluated?

Response:

Thanks for the comment. The German Ultrafine Aerosol Network (GUAN) includes 17 stations. However, one station Leipzig-West (LWE) has been terminated in 2016 and it shows high similarity as site Leipzig-TROPOS (LTR). Therefore, we decided not to include LWE in this study. Only 16 sites were used. To avoid confusion, we have deleted LWE in site description and in Fig.1, table 1 and 2 in the manuscript.

At site LAN, there is no measurement of eBC mass concentration. Thus, we decided to use another site Raunheim which is not a GUAN site but close to LAN, to help detect the BC trend. In the revised manuscript, we have deleted the results from Raunheim to avoid confusion.

For the question about the number of trends, as shown in Fig. 2 in the manuscript, the eBC mass concentrations were not measured at MST and LAN. PNSD measurements at MST and ZSF start from diameter of 14 and 20 nm, respectively. Thus no information of $N_{[10\text{-}30]}$ is available at these two sites. Therefore, for 16 sites and five parameters, only 76 trends in total were evaluated. To make it clear, a new table (Table S1) has been added in the supplemental material giving the number of analysed stations with respect to all five parameters, and the sentence in Sect. 3.1 has been revised as “For the five parameters at the 16 sites (76 trends in total, see Table S1 in SM), the trends…”

Table S1: Number of sites used in trend analysis.

Parameters	Number of stations analysed	Excluded stations
eBC	14	MST, LAN
$N_{[20–800]}$	16	--
$N_{[10–30]}$	14	MST, ZSF
$N_{[30–200]}$	16	--
$N_{[200–800]}$	16	--

(4) For the PNC data description, the uncertainty of the PNC measurements could be discussed in the text a bit more, (L164 etc.) and compared to the presented trends.

Response:

Thanks for your comment. The following paragraph has been added in Sect. 2.2 in the text.

“Quality assurance of PNSD measurements in GUAN are periodically done to ensure that measurements remain stable both instrument to instrument (or site to site) and instrument to standard. Monthly maintenance and onsite/laboratory inter-comparisons with a reference MPSS with a frequency between one to four times per year as recommended by Wiedensohler et al. (2018) are done by the World Calibration Centre for Aerosol Physics (WCCAP, http://www.wmo-gaw-wcc-aerosol-physics.org/). These procedures can ensure an accuracy of ±10 % for PNCs over the entire measurement period (Birmili et al., 2016). Although the uncertainty of PNCs is comparable or higher than their annual trends (Sect. 3.1), with the application of periodical quality assurance procedures, there should be no monotonicity change or systematic bias in the measurement uncertainties. Therefore, the influence of the measurement uncertainty on the detection of long-term trends of PNCs is assumed to be negligible.”

(5) In the 5.1 Section, a mean value of meteorological parameters is used for all stations. Would not it be better to have at least three different averages for the different types of stations? It would be difficult to compare one T and RH value for Alpine site, city etc. (L416)

Response:

Thanks for the comments. Following your suggestion, the 76 DWD stations were classified into three categories (Table 5 in the manuscript): urban background, regional background and mountain area. And section “4.1 Influence of precipitation, temperature, wind speed on the detected trends” has been revised accordingly.

Table 5: Trends of meteorological parameters for the three site categories in Germany. The bold numbers are the statistically significant slopes at the 95 % significance level. The daily meteorological data are from Germany's National Meteorological Service (Deutscher Wetterdienst, DWD).

season		Urban background	Regional background	Mountain area
Spring	Precipitation mm $year^{-1}$ (% $year^{-1}$)	−0.02 (−1.0)	0.00 (0.0)	−0.02 (−0.5)
	Temperature °C $year^{-1}$	−0.04	−0.03	−0.02
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	0.01 (0.2)	0.02 (0.3)	0.04 (0.6)
Summer	Precipitation mm $year^{-1}$ (% $year^{-1}$)	**−0.14 (−5.5)**	**−0.15 (−5.8)**	**−0.20 (−4.7)**
	Temperature °C $year^{-1}$	**0.15**	**0.13**	**0.16**
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	0.00 (0.0)	0.02 (0.4)	**−0.08 (−1.4)**
Autumn	Precipitation mm $year^{-1}$ (% $year^{-1}$)	−0.07 (−3.9)	−0.05 (−2.5)	**−0.07 (−1.9)**
	Temperature °C $year^{-1}$	**0.37**	**0.36**	**0.29**
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	−0.02 (−0.8)	−0.01 (−0.3)	**−0.09 (−1.2)**
Winter	Precipitation mm $year^{-1}$ (% $year^{-1}$)	0.02 (1.3)	0.04 (1.8)	**0.14 (3.1)**
	Temperature °C $year^{-1}$	**0.41**	**0.43**	**0.34**
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	0.02 (0.5)	0.05 (0.9)	**0.13 (1.5)**

(6) Also the 5.2 section needs more detailed methodology description. Why 15 clusters were used, what data were used for trajectory calculation? And mainly, why the analyses have not been done for the whole period? No changes in the period 2009-2014 do not automatically mean there will be no changes in 2009-2018 as well (L444). also it is not described what is the difference between for example A1 and A2 cluster?

Response:

Thanks for the comment. Following your suggestion, we have added a more detailed description in supplementary information (Sect. 3 in the supplemental material) about the back-trajectory cluster method, including the basic description, data sources, cluster algorithm, and the evaluation of cluster results.

Guided by experience from previous studies (Engler et al., 2012; Ma et al., 2014), we tested the cluster algorithm for a range of cluster numbers *k* between 8 and 19. The best solution was obtained with cluster number 15. More and more redundancies in the cluster composition (i.e. cluster means close to each other) were observed for $k > 15$; while reducing the number of clusters below 15 would, conversely, merge clusters that could be clearly identified as different typical weather situations in Central Europe.

The trajectories were calculated using a PC version of HYSPLIT (Stein et al., 2015) with Global Data Assimilation System (GDAS) analysis set which provides meteorological fields every 3 hours, at a horizontal resolution of 1°, and at numerous standard pressure levels. In our Back Trajectory and Temperature Profile (BTTP) cluster method, vertical profiles of pseudopotential temperature θ_v retrieved from radiosounding data at seven stations are also used for the classification of trajectories.

We have extended the clustering for the whole time period (2009−2018) in the revised manuscript. The corresponding figures, tables and text have been updated. The new results are similar as that for the period of 2009−2014.

For the comment "also it is not described what is the difference between for example A1 and A2 cluster?", following description of the 15 air mass types has been added in Sect. 4.2 in the text.

"The 15 air mass types are named by seasons (CS: cold season; TS: transition season; and WS: warm season) and synoptic patterns (ST: Stagnant; A1: Anti-cyclonic with air mass originating from Eastern Europe; A2: Anti-cyclonic with air mass originating from west; C1: cyclonic with air mass originating from relatively south; C2: cyclonic with air mass originating from the north). Table 6 lists the basic statistical information of the 15 air mass types."

Minor comments:

(1) The manuscript would definitely profit from a native speaker check, there are multiple not very usual English phrases –

L60 early regions (first?),

L343 declined emissions => decreased?,

L493+506 downward trend => decreasing trend?

L330+331 LENGTH of the time series - sometimes a verb is missing (L277 monthly median time series WERE USED?, L282 only MEL SHOWS increase?) or mismatched (L355 there is no difference can be seen between, L440 shown => showed?)

there is a superfluous use of commas, for example as "it should be noted that, three sites: L300, L312,335 etc",

also some minor typos, for example L316 concentrations are in consistent, L321, Po Valey, one large industrial district.

Response:

Many thanks for the corrections and suggestions. Most parts of the manuscript have been rewritten, and above sentences have been corrected or deleted. The manuscript has been also edited by Elsevier Language Editing Services to improve the language.

(2) In the equation description in 2.3.2 and 2.3.3. sections, the symbols are not clearly described, so it is quite difficult to follow the methodology. For example, Eq. 3 does not say what the 2 pi t or 4 pi t means etc. on the other end, it is not necessary to show how a general vector or matrix looks like, L219 to 221 (with vector missing the C). If the description would be less technical and more explaining what is what, it would be easier to follow. Similarly, the theta in Eq. 9 is not explained at all, and although I know how the signum function works, I cannot recognize it in the eq?

Response:

Thanks for the comment. The GLS/ARB is a well-developed method and adapted by Asmi et al. (2013) for trend analysis of aerosol concentrations. Since we use exactly the same method as that in Asmi et al. (2013) and all details can be found in the book by Mudelsee (2010) and the paper by Asmi et al. (2013), we decided not to repeat the details in the main text, to make the text shorter and easy to read. Section "**2.3.2 Generalized least-square-regression and auto-regressive bootstrap confidence intervals**" and "**2.3.3 Regional Mann-Kendall test**" has been shortened accordingly.

(3) L363+368 eBC instead of BC?

Response:

Thanks. We have rewritten the entire paragraph and this sentence has been removed. In the revised manuscript, the term "BC" is used to stand for aerosol species black carbon, and the term "eBC" is used to stand for the measurement data of MAAP and Aethalometer.

(4) L72-73 sentence does not fit either to the preceding or the following sentences

Response:

The sentences have been revised as "For domestic heating emission, the unsuitable fuels are listed and their emission values are defined. For traffic emission, low emission zones (LEZs) were set up to limit the emission of nitrogen oxide and aerosol particles from traffic exhaust."

(5) What does it mean "dataset is sufficient for true slopes"? L336

Response:

The section has been moved to supplementary information. And the sentence in the main text has been revised as "Robustness analysis (see Sect. 1 in the supplementary material) suggests that the time span of our dataset is long enough for slope detection and that the influence of measurement uncertainty is negligible.”

(6) L366 higher reduction rate is observed when human activities are more intensive?

Response:

Here, “human activities” means traffic, domestic heating, cooking etc. This section has been rewritten and the corresponding sentence has been removed.

(7) Why is the N10-30 described as “influenced by NPF” called young Aitken and not nucleation as usual?

Response:

The term “young Aitken mode” has been replaced by “nucleation mode” in the revised manuscript.

(8) Number of references at some sections is redundant, for example 8 references stating non-parametric test are used for trend analysis? L183

Response:

Thanks. Some of the references have been removed in the manuscript.

(9) L398 do you expect the biogenic emissions in summer to have a trend? If not, they should not mask the anthropogenic trend?

Response:

We do not except any trend in the biogenic emission in summer. Biogenic emissions contribute considerable SOA precursors and thus a large contribution on PNCs. Therefore, the relative contribution of anthropogenic emission is lower in summer than in other seasons. Without any strong long-term variation in biogenic emission (means a large portion in PNC does not change), its stable contribution on PNCs may lower the relative decreasing rates in PNCs in summer. To make it clearer, the sentence has been revised as:

“Other than the low residential emission in warm seasons, another reason might be the strong seasonal variations in biogenic emissions (Asmi et al., 2013). Biogenic emissions contribute considerable secondary organic aerosol (SOA) precursors in summer and thus a higher contribution on PNCs. Without any strong long-term variation, the stable contribution of biogenic emissions on PNCs might lower the relative decreasing rates in PNCs in summer.”

(10) L526 the a) explanation does not explain anything, it just repeats the previous sentence?

Response:

Thanks. We have rewritten the entire paragraph and this sentence has been removed.

Reference

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Decreasing Trends of Particle Number and Black Carbon Mass Concentrations at 16 Observational Sites in Germany from 2009 to 2018

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Abstract~~. Anthropogenic emissions are a dominant contributor to air pollution. Consequently, mitigation policies have attempted to reduce anthropogenic pollution emissions in Europe since the 1990s. To evaluate the effectiveness of these mitigation policies, the German Ultrafine Aerosol Network (GUAN) was established in 2008, focusing on black carbon and sub-micrometer aerosol particles, especially ultrafine particles. In this investigation, trends of the size-resolved particle number concentrations (PNC) and the equivalent black carbon (eBC) mass concentration over a 10-year period (2009-2018) were evaluated for 16 observational sites for different environments among GUAN. The trend analysis was done for both, the full-length time series and on subsets of the time series in order to test the reliability of the results. The results show generally decreasing trends of both, the PNCs for all size ranges as well as eBC mass concentrations in all environments, except PNC in 10-30< 200 nm at regional background and or mountain sites. The annual slope of the eBC mass concentration varies between -13.1 % and -1.7 % per year. The slopes of the PNCs vary from -17.2 % to -1.7 %, -7.8 % to -1.1 %, and -11.1 % to -1.2 % per year (only statistical significant trends) for 10-30 nm, 30-200 nm, and 200-800 nm particle diameter, respectively.The annual slope of the eBC mass concentration varies between -7.7 % and -1.8 % per year. The slopes of the PNCs varies from -6.3 % to 2.7 %, -7.0 % to -2.0 %, and -9.5 % to -1.5 % per year (only significant trends) for 10-30 nm, 30-200 nm, and 200-800 nm particle diameter, respectively. The regional Mann-Kendall test yielded regional-scale trends of eBC mass concentration, $N_{[30-200]}$ and $N_{[200-800]}$ of -5.03.8 %, -2.0 5 % and -2.4 9 %, respectively, indicating an overall decreasing trend for eBC mass concentration and sub-micrometer PNC (except $N_{[10-30]}$) all over Germany. The decreasing trends are found on the total emission of BC, $PM_{2.5}$ and precursors, suggesting the reduced anthropogenic emission is the dominant factors for the reduced eBC mass concentration and PNCs in Germany. The most significantstrongest decrease was observed on working days and during daytime~~

~~in urban areas, which implies a strong evidence of reduced anthropogenic~~traffic ~~emissions. For the seasonal trends, stronger reductions were observed in winter~~ due to limited emission from domestic heating~~. Possible reasons for this reduction can be the increased average ambient temperatures and wind speed in winter, which resulted in less domestic heating and stronger dilution. In addition, decreased precipitation in summer also diminishes the decrease of the PNCs and eBC mass concentration. For the period of interest, there were no significant changes in long-range transport patterns. The most likely factors for the observed decreasing trends are declining anthropogenic emissions due to emission mitigation policies of the European Union.~~

Anthropogenic emissions are ~~a~~ dominant contributors to air pollution. Consequently, mitigation policies have been attempted since the 1990s in Europe to reduce pollution by anthropogenic ~~pollution~~ emissions~~in Europe since the 1990s~~. To evaluate the effectiveness of these mitigation policies, the German Ultrafine Aerosol Network (GUAN) was established in 2008, focusing on black carbon (BC) and sub-~~micrometer~~micrometre aerosol particles~~the~~. In this study, long-term trends of atmospheric particle number concentrations (PNCs) and equivalent ~~black carbon~~BC (eBC) mass concentration over a 10-year period (2009~~-~~–2018) were determined for 16 GUAN sites ranging from roadside to high Alpine environment~~s,~~. Overall, statistically significant ~~downward~~decreasing trends are found for most of these parameters and environments in Germany. The annual relative slope of eBC mass concentration varies between ~~-~~–13.1 % and ~~-~~–1.7 % ~~yr-1~~per year. ~~And t~~The slopes of the PNCs vary from ~~-~~–17.2 % to ~~-~~–1.7 %, ~~-~~–7.8 % to ~~-~~–1.1 %, and –11.1 % to ~~-~~–1.2 % ~~yr-1~~per year for 10~~-~~–30 nm, 30~~-~~–200 nm, and 200~~-~~–800 nm size ranges, respectively~~of micrometre~~. The reductions ~~of~~in various anthropogenic emissions are found to be the dominant factors responsible for the decreasing trends of eBC mass concentration and PNCs. The diurnal and seasonal variations ~~of~~in the trends clearly show the effects ~~of for~~of the mitigation policies ~~of~~for road transport and residential emissions. The influences of other factors such as air masses, precipitation, and temperature ~~etc.~~ were also examined and found to be less important or negligible. This study proves that a combination of emission mitigation policies can effectively improve the air quality on large spatial scales. It also suggests that a long-term aerosol measurement network ~~with~~in multi-type sites is an efficient and necessary tool for ~~the verification~~evaluating emission mitigation policies.

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1 Introduction

Epidemiological studies have shown that increased particulate air pollution due to anthropogenic emissions leads to adverse health effects ~~upon health, including not only respiratory but also cardio-vascular disease~~ (Seaton et al., 1995)~~, further~~ and increases global disease burden (Cohen et al., 2017). Among the ambient sub-~~micrometer~~micrometre aerosol particles (diameter < 1 µm), ultrafine particles (UFP, diameter < 100 nm) share the ~~greatest~~ largest number fraction ~~of particles~~. The ~~A main rationale for~~ UFP-driven effects upon health is their ~~ability to~~ can penetrate deep into lungs and translocate to other organs such as brain, caus~~e~~ing ~~other~~ health problems such as respiratory and cardiovascular diseases (Kreyling et al., 2006; Schmid and Stoeger, 2016). ~~Previous studies suggested that exposure to UFP might lead to an increased probability of health hazards (Kreyling et al., 2006; Schmid and Stoeger, 2016), although at present, epidemiological evidence for~~ of ~~their effects upon human health remain~~ insufficient ~~mixed due to a number of reasons (Ohlwein et al., 2019). A main rationale for UFP-driven effects upon health is their ability to penetrate deep into lungs and translocate to other organs such as brain, cause~~

~~other health problems such as respiratory and cardiovascular diseases (Kreyling et al., 2006; Schmid and Stoeger, 2016).~~ In urban areas, ~~a significant fraction of UFP mass consists of~~ black carbon (BC) produced by incomplete combustion of fossil fuel and biomass accounts for a significant fraction of UFP mass~~, which is produced due to incomplete combustion of fossil fuel and biomass and then released to the atmosphere~~ (Chen et al., 2014; Cheng et al., 2013; Pérez et al., 2010). ~~Since~~ As BC ~~may~~ could operate as a universal carrier of a wide variety of toxins such as polycyclic aromatic hydrocarbons (PAH) and heavy metals into ~~the~~ human body, exposure to BC ~~shows~~ could cause acute health effects such as ~~strong relevance with~~ cardiopulmonary morbidity and mortality (Janssen et al., 2012).

~~To reduce the harmful effects of air pollution, To reduce the harmful effects caused by air pollution, emission mitigation policies were implemented around the world. T~~tThe European Union (EU) ~~is~~ was one of the ~~early earliest~~ earliest ~~first~~regions~~,~~ around the world to implement~~where~~ing emission reduction policies to reduce the harmful effects of air pollution ~~have been implemented~~. ~~The main policy instruments on air pollution within the EU include the Ambient Air Quality Directives and the National Emission Ceilings Directive.~~ EU emission mitigation legislations are directly formulated based on emission sources~~, such as road transport, industry and residential~~. In Europe, the main anthropogenic sources ~~to~~ of primary aerosol particles are fuel combustions from industrial installations (power generation, industry, etc.), non-road and road transport, and domestic heating ~~etc.~~ (European Environment Agency, 2017). The Member States of EU were required to ~~draw up a~~draft National Programmes to the Commission (http://ec.europa.eu/environment/air/reduction/implementation.htm). ~~For example~~~~example, i~~In Germany, the Federal Environment Ministry issued the Federal Emission Control Regulations (~~German:~~ Bundes-Immissionsschutzverordnung, BImSchV). To reduce the emission from industrial installations, the BImSchV ~~requires~~ regulates~~the~~ permits for~~of~~ construction and operation of~~for~~ ~~some~~ industrial installations~~, in accordance with the Federal Emission Control Act~~. The emission limits for ~~from~~ large combustion, such as for that from power plants, are defined as well. For domestic heating emission, the unsuitable fuels are listed and their emission values are defined. For ~~d to control the emission.~~ traffic emission~~In Europe, traffic emissions have been found to be a dominant contributor to air pollution in the urban outdoor atmosphere (Kumar et al., 2010; Pey et al., 2009). A~~, low emission zones (LEZs) were set up ~~nother policy, the clean air plan (German: Luftreinehalteplan), has great practical importance on the operation of vehicles. It set up low emission zones (LEZs) in Germany~~ to limit the emission of nitrogen oxide and aerosol particles from ~~the~~ traffic exhaust. ~~Previous short-term studies indicated that a LEZ can reduce the pollutant concentration immediately after its implementation, as a result of the absence of the most polluting vehicles (Rasch et al., 2013; Qadir et al., 2013; Jones et al., 2012).~~

It is important to ~~To~~ evaluate the effectiveness of the implemented ~~those~~ emission mitigation policies~~,~~ through~~.~~ ~~And~~ long-term observations of pollutants ~~are crucial, especially for those health-related pollutants,~~ such as sub-~~micrometer~~micrometre particles and BC ~~are the only effective way~~. There have been many studies ~~about~~ on the long-term trends of particle number concentration (PNC) and/or BC mass concentration since the 1990s. These studies have concluded that emission mitigation policies may reduce ~~the~~ human exposure to the pollutants~~,~~ and the results were important for the policy makers (Barmpadimos et al., 2011; Masiol et al., 2018; Kutzner et al., 2018; Putaud et al., 2014~~; Sabaliauskas et al., 2012; Wang et al., 2012~~). ~~However, m~~Most of these studies were conducted at roadside or urban background, which are largely dominated by traffic emissions. ~~Only a few~~ ~~Only a f~~Few of ~~them~~~~studies~~ have focused~~inged~~ on long-term trends ~~of the PNC or the BC mass concentrations~~ at the regional background setting (Asmi et al., 2013; Barmpadimos et al., 2011; Murphy et al., 2011). Murphy et al. (2011) found that between 1990 and 2004, the elemental carbon (EC) mass concentration decreased in several of the national

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parks and other remote sites in the US~~ between 1990 and 2004~~. This result was an indication that the emission control policies were effective in reducing the EC mass concentration in the background air across the US. Asmi et al. (2013) analysed the long-term change of PNC at the regional background and remote sites in Europe, North America, Antarctica, and Pacific Ocean islands during 2001–2010. The results showed that decreased PNCs could likely be explained by the reduction of anthropogenic emissions. ~~However, However, tare oney the long-term trend studies of PNC and BC mass concentration measured in parallel and covering different environments from roadside, urban background to regional background and remote areas in the same region have not been done.~~~~For example,~~ Kutzner et al. (2018) evaluated the ~~long-term~~ trend of BC over Germany ~~including industrial,~~based on measurement at ~~rural,~~ traffic, ~~ic and~~ urban background, and rural sites ~~during~~infor the period of 2005~~ –~~-2014~~. The result~~, and concluded that the observed decreasing trends in BC are likely owing largely to mitigation measures in the traffic sector. However, there is still a lack of a thorough investigation of the connections between the long-term trends of PNCs/BC and the change of different anthropogenic emissions. A better understanding of the influence of the inter-annual variation of meteorological conditions on the observed trends is also needed.~~confirminged that the emission control policy in the last two decades haves most likely contributed to the mitigation ofe BC mass concentration in Germany and Europe. However, the long-term trend for both BC and PNC considering various site conditions from traffic, urban, regional background and high altitudes for the time period after 2010 have yet not been done.~~~~However, the long-term trend studies of PNC and BC mass concentration measured in parallel and covering different environments from roadside, urban background to regional background and remote areas in the same region have not been done.~~

Based on a unique dataset ~~of~~from the German Ultrafine Aerosol Network (GUAN), ~~T~~this study investigates the long-term variation ~~of~~in the regional PNC and BC mass concentration, ~~in order~~ to ~~takes Germany as an example to~~ understand the effectiveness of the emission mitigation policies ~~on the~~in reducingt~~ion~~ the~~of~~ PNC and BC in Germany~~ the regional PNC and BC mass concentration~~. ~~In this investigation , trend analysis was done for the sub-micrometer PNC (diameter < 1 µm) and the equivalent black carbon (eBC) mass concentration in Germany based on a unique dataset of the German Ultrafine Aerosol Network (GUAN).~~ The study was conducted for~~For~~ the period of ~~study (~~2009–2018~~),~~ with data from 16 observational sites representing different types of environment (roadside, urban background, regional background, low mountain, and high Alpine)~~ have been included in this study, ranging from roadside to high Alpine~~. The overall, ~~weekly,~~ diurnal, and seasonal trends of PNCs and BC~~and the robustness of the trend~~ ~~were~~ are evaluated~~.~~ ~~a~~And the role of potential decisive factors, including not only emission mitigation policies, but also other potential drivers (i.e. inter-annual change in~~the~~ meteorological conditions ~~change~~ and long-range transport patterns~~ change~~) are discussed.

~~To determine, if the past emission mitigation policies are the decisive factor for the long-term trends of the eBC mass concentration and PNC at different environments, the annual change of total emission of BC, $PM_{2.5}$ and precursors were compared with the change of measured parameters. Moreover, the effect of LEZ was analysed to illustrate how the emission mitigation policy can effectively reduce the PNC and eBC mass concentration in the urban area. At last, thethe influences of other potential drivers (i.e. the meteorological condition change and long-range transport pattern change) are also evaluated to discuss their impact on the long-term trend of measured parameters in GUANed.~~

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2 Data~~set~~ and methods

2.1 The German Ultrafine Aerosol Network (GUAN)

The ~~German Ultrafine Aerosol Network (GUAN)~~GUAN is a specialized network in Germany that provides~~ing~~ continuous measurements including sub-micromet~~er~~re particle number size distribution (PNSD) and the equivalent BC (eBC) mass concentration in diverse environments including ~~,~~roadside, urban background, regional background, low mountain range and high Alpine. ~~The sites investigated in this study belong to GUAN, which~~ The GUAN combines federal and state air quality monitoring stations~~,~~ as well as atmospheric observatories from research institutes, aiming at a better understanding of sub-~~micrometer~~ micrometre PNC and BC with respect to human health and climate impact (Birmili et al., 2016). ~~GUAN is a specialized network in Germany, which provides continuous measurements including sub-micrometer particle number size distribution (PNSD) and eBC mass concentration, with diverse environments from roadside, urban background, regional background, low mountain range to high Alpine.~~

Table 1 lists the basic information of the GUAN sites evaluated in this study. The locations of ~~GUAN~~ all 16 sites are illustrated in Fig. 1. A ~~summarized~~ summarised description of ~~GUAN~~ the sites is given here ~~with~~ and more details are available in Birmili et al. (2016). Among the ~~17~~ 16 sites, ~~eight~~ seven are located in the state of Saxony: Leipzig-Mitte (LMI), Leipzig-Eisenbahnstraße (LEI), Leipzig-~~-~~Leibniz Institute for Tropospheric Research (TROPOS)~~TROPOS~~ (LTR), ~~Leipzig-West (LWE),~~ Melpitz (MEL), Dresden-Nord (DDN), Dresden-Winckelmann-straße (DDW), and Annaberg-Buchholz (ANA). LMI and LEI are two roadside stations in Leipzig. LMI~~The former one~~ is located ~~at~~ on roadside in an open area in the city cent~~er~~re, while LEI ~~the latter one~~ is a street canyon station. The traffic volumes at these two sites are 44~~-~~000 and 12~~-~~000 vehicles per day, respectively. ~~LTR and LWE are urban background sites located in the city of Leipzig with 10 km apart.~~ LTR is an atmospheric research station situated on the roof of~~operated by~~ the ~~Leibniz Institute for Tropospheric Research (TROPOS)~~ main building of TROPOS~~. The station is situated on the roof of the TROPOS institute building~~. ~~LWE was is located in a park on the premises of a hospital, with negligible traffic influence.~~ Station MEL ~~operated by TROPOS since 1992,~~ is located in a farmland about 50 km from Leipzig. Previous studies have showed that MEL can well represent the regional background atmosphere of Central Europe (Spindler et al., 2013). Two stations are located in the city of Dresden: a roadside ~~station~~ site DDN with ~~the~~ a traffic volume of ~~∼~~about 36~~-~~000 vehicles per day~~,~~ and an urban background site DDW,~~, which is~~ 1.7 km away from the city centre. ANA is an urban background station ~~for Saxon State Office for Environment, Agriculture and Geology (LfULG),~~ located in the city of Annaberg-Buchholz in the Ore mountain area~~,~~ about 10 km away from the German-Czech border (Schladitz et al., 2015).

Three stations are located in the lowlands of ~~the~~ Northern Germany: Bösel (BOS), Neuglobsow (NEU) and Waldhof (WAL). The urban background site BOS is located in the village of Bösel, about 100 km from the North Sea~~, i~~. It is, ~~th~~therefore,~~us~~ partly influenced by maritime air masses. NEU and WAL are located in forests, representing regional background conditions of~~in~~ the Northern Germany lowlands.

Two stations, Langen (LAN) and Mülheim-Styrum (MST), are located in the western part of Germany. LAN is an urban background site located in the city of Langen~~,~~ at the edge of a residential area and a forest. Emissions from the Frankfurt's Rhein-Main airport (about 5 km to the southeast) ~~may~~ might sometimes influence the observations at LAN. MST is situated in the western end of the Ruhr area, — the largest urban cluster area in Germany.

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Four stations are located in the southern part of Germany, including one urban background site Augsburg (AUG), two low mountain range sites~~,~~ Schauinsland (SCH) and Hohenpeißenberg (HPB), and one high Alpine site Zugspitze (ZSF~~, Schneefernerhaus~~). AUG is located on the premises of Augsburg's University of Applied Sciences, about 1 km southeast of Augsburg city ~~center~~centre. The two low mountain range sites SCH and HPB are surrounded mainly by forests and agricultural pastures, and are located at the~~with~~ ~~. Their~~ elevations ~~are~~ of 1205 and 980 m above sea level (a.s.l.~~.~~), respectively. The high Alpine site ZSF is located at 2670 m a.s.l~~.,~~. and 300 m below the summit of the Zugspitze, at the south side of the highest mountain in Germany.

~~It needs to be noted that the selection of sites in GUAN could not be designed from scratch. As financial resources to perform specialized air pollution measurements are limited, GUAN has incorporated such sites where sub-micrometer particles were already measured by one of the partner institutions, or sites that could be co-established with the aid of other research projects or programs. This explains the incomplete geographic coverage of Germany with GUAN measurement sites.~~

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2.2 Instrumentation

The technical details of the PNSD and the eBC mass concentration measurements at each GUAN site~~s~~ are summari~~z~~sed in~~ this section and~~ Table 2.~~ Details~~ More detailed information ~~of~~ on the instrumentation and data processing ~~techniques~~ are provided in Birmili et al. (2016). Depending on individual set-up, ~~the~~ PNSD are measured either by the Mobility Particle Size Spectrometers (MPSS, Wiedensohler et al., 2012) or by the Dual Mobility Particle Size Spectrometers (D-MPSS). ~~Regenerative Nafion~~~~The a~~Aerosol dryers are used to dry the aerosol sample to a relative humidity below 40 % (Swietlicki et al., 2008). The PNSD is ~~obtained~~ retrieved ~~from~~ from the raw mobility distribution~~s~~ ~~by~~ using an inversion algorithm (Pfeifer et al., 2014), including the commonly used bipolar charge distribution (Wiedensohler, 1988). ~~Corrections~~ The corrections for ~~diffusional~~ particle losses in instruments and inlets ~~were~~ are made ~~according to~~based on Wiedensohler et al. (2012). ~~Due to the individual settings of MPSS at GUAN sites, the quality of the PNSD was ensured by onsite or laboratory inter-comparisons conducted by the World Calibration Centre for Aerosol Physics (WCCAP, http://www.wmo-gaw-wcc-aerosol-physics.org/) at TROPOS. The frequency of quality control is between one to four times per year, as recommended by Wiedensohler et al. (2018).~~

Mass concentrations of eBC ~~have been~~are measured ~~by~~ using the Multi-Angle Absorption Photometers (MAAP, Thermo Scientific, model 5012), except ~~in~~ for AUG, where an Aethalometer (Type 8100, Thermo Fisher Scientific Inc.) is used. For the MAAP measurement, eBC mass concentration is obtained using a mass absorption cross section of 6.6 m^2 g^{-1} ~~for~~ at the wavelength of 637 nm (Petzold and Schönlinner, 2004; Müller et al., 2011). ~~No~~ eBC data ~~are~~ is not available for LAN and MST.

Quality assurance of PNSD measurements in GUAN are periodically done to ensure that measurements remain stable both instrument to instrument (or site to site) and instrument to standard. Monthly maintenance and onsite/laboratory inter-comparisons with a reference MPSS with a frequency between one to four times per year as recommended by Wiedensohler et al. (2018) are done by the World Calibration Centre for Aerosol Physics (WCCAP, http://www.wmo-gaw-wcc-aerosol-physics.org/). These procedures can ~~The quality assurance of measurements were done in GUAN to ensure that the measurement remain stable both site to site and instrument to reference, including annual/monthly maintenance and comparisons to reference instruments. The aim of the quality assurance is to~~ ensure an accuracy of ~~within a few percent for the eBC mass concentration measurements, of a few percent for the PNSD, and of~~ ±10 % for PNCs over the entire measurement period (Birmili et al., 2016).

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Although the uncertainty of PNCs is comparable or higher than their annual trends (Sect. 3.1), with the application of periodical quality assurance procedures, there should be no monotonicity change or systematic bias in the measurement uncertainties. Therefore, the influence of the measurement uncertainty on the detection of long-term trends of PNCs is assumed to be negligible.

To condense the information provided by PNSD, ~~we chose~~ three particle size ranges, 10−-30 nm, 30−-200 nm, and 200−-800 nm, are chosen ~~es~~ to obtain integrated PNCs~~: 10-30 nm, 30-200 nm, and 200-800 nm~~. ~~The young Aitken mode~~ $N_{[10-30]}$ represents the ~~Aitken~~nucleation mode ~~the~~ in which particles are freshly formed by homogeneous nucleation from either photochemical processes or downstream of traffic exhausts. ~~Aitken mode particles~~ $N_{[30-200]}$ represents the Aitken mode in which particles are either directly emitted from incomplete combustion or grown by condensational growth. ~~The accumulation mode~~ $N_{[200-800]}$ represents the accumulation mode in which ~~aged~~ particles~~, which~~ have ~~underwent~~ undergone condensational growth or cloud processing during ~~long-range~~ transport. ~~Since~~ As the particles below 20 nm ~~were~~ are not measured ~~all the time~~ from 2009 to 2018 at ZSF and MST, we use $N_{[20-800]}$ to represent total PNC in this study instead of $N_{[10-800]}$. ~~Data coverage can largely influence the evaluation of long-term trends.~~ Figure 2 illustrates the time span for which data ~~coverages of~~~~were~~are available at the 16 stations in GUAN ~~until the end of~~~~from~~during the study period of 2009 to 2018~~, except LWE. LWE is not evaluated in this study since its observation shows high similarity with LTR (Sun et al., 2019) and it was terminated at the end of 2016~~. ~~To~~ As a sum-up, Table S1 in the ~~Sumpplement~~Supplemental Material (SM) gives ~~the selected~~ the number of stations ~~and measured parameters which are~~ used in ~~following~~ trend analysis~~the~~.

2.3 Trend analysis methods

~~Most of the e~~Environmental data are usually not normally distributed. Therefore, non-parametric methods are often used to detect the long-term trends (e.g., Asmi et al., 2013;~~ Barmpadimos et al., 2011;~~ Bigi and Ghermandi, 2014; ~~Collaud Coen et al., 2007;~~ Collaud Coen et al., 2013~~; Mejía et al., 2007; Murphy et al., 2011; Sharma et al., 2006~~). Detection of long-term~~,~~ linear trends ~~might~~ can be affected by several factors~~,~~ such as ~~the~~ time span and time resolution ~~of available data~~, ~~the~~ magnitude of variability, autocorrelation and periodicity of~~in~~ the time series (Weatherhead et al., 1998). To analyse the ~~temporary~~ long-term trends of the PNCs and the eBC mass concentrations, two trend evaluation methods, ~~C~~a customi~~z~~sed Sen-Theil trend estimator and ~~G~~the generalized ~~L~~least-~~s~~Square-regression (GLS), were used in this study. Moreover, the r~~R~~egional Mann-Kendall test was used to detect the overall trends over the whol~~l~~e study region ~~studied~~. To ensure the comparability of trend slopes among the different sites, relative slopes (absolute slope divided by the median value of the whole time series) in % per year were used.

2.3.1 ~~Customized~~ Customised Sen-Theil trend estimator

~~The c~~The cu~~u~~stomi~~z~~sed Sen-Theil trend estimator (~~customized~~ customised Sen's estimator, hereafter) is a modified non-parametric procedure based on the ordinary ~~normal~~ Sen's slope estimator~~, regardless the influence of outlier, missing values and statistical distribution~~ (Sen, 1968; Theil, 1992; Birmili et al., 2015). This approach ~~estimates~~ can give the true slope of atmospheric parameters by fully considering the effect of their ~~some~~ periodic variations, ~~of atmospheric pollution,~~ such as, seasonal, weekly, ~~or~~ and diurnal cycles, and avoid the influence of outliers, missing values, and statistical distribution of the data. ~~It is thus possible to estimate the true slope by this approach for a the shorter~~small ~~data set with higher time resolution, for example, a 5~~five~~-year hourly time series.~~ Based on

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the hourly or daily time series $x(i)$, firstly, the rates of change $m_{i,k}$ ~~on~~ of each data pair $[x(i), x(i + k \times 364\ days)]$ is calculated as~~:~~

$$\cancel{m_{i,k} = \frac{(x(i+\Delta t)-x(i))}{\Delta t}} \quad \begin{cases} m_{i,k} = \frac{(x(i+\Delta t)-x(i))}{\Delta t} \\ \Delta t = k \times 364\ days \end{cases} \qquad (1)$$

~~with $\Delta t = k \times 364\ days$.~~

where k is the integer. Δt ensures that each data point ~~can be~~is ~~only~~ compared only with data points that are separated by a multiple of 52 weeks (~~=~~364 *days*), that is, ~~two~~ data points from two different years are compared only if they both were measured on~~belong to~~ the same hour of the day, day of the week, and season of the year. For our dataset~~each time series~~, ~~some~~ more than 10000 slopes $m_{i,k}$ are calculated for each time series. The median of those slopes is taken as the true slope ~~m over~~for the whole period. Significance and confidence interval (CI) of the trends are determined at 95 % confidence level from the distribution of $m_{i,k}$.

2.3.2 Generalized ~~Least~~least-~~Square~~square-regression and ~~Auto~~auto-~~Regressive~~ regressive ~~Bootstrap~~ bootstrap confidence intervals ~~(GLS/ARB)~~

The second method used to detect the trends is the generalized least-square-regression (~~Generalized Least-Square-regression (~~GLS~~)~~ ~~(~~;) ((Mudelsee, 2010; Asmi et al., 2013). A brief introduction of the method is given here, for details refer to Mudelsee (2010) and Asmi et al. (2013). ~~The~~ GLS is an approach for estimating the linear parameters in a linear regression model.

For a time series of an ~~observation~~observed parameter $x(i)$, compactly written as $\{t(i), x(i)\}_{i=1}^{n}$, ~~we separate the time series~~~~define~~ the linear regression model can be defined as:

$$x(i) = \beta_1 + \beta_2 t(i) + \Omega(t(i)) + S(i)e(i) \qquad (2)$$

where β_1 and β_2 are the two trend parameters (intercept and slope~~),~~). $\Omega(t(i))$ is a seasonal component. $S(i)$ is ~~the~~ a variability function scaling the random noise term $e(i)$~~., $\Omega(t(i))$ is the seasonal component (Asmi et al., 2013, Sect. 2.5,). In this study, four seasonal components are defined as:~~

~~= sin(), = sin(),~~

~~= cos(), = cos(). (3)~~

The GLS regresses ~~two~~ the trend and ~~four~~ seasonal parameters (denoted as $\boldsymbol{\beta}$, ~~thereafter~~hereinafter) by minimizing the sum of squares of the residuals ($SSQG$)~~:~~

$$SSQG(\boldsymbol{\beta}) = (\boldsymbol{x} - \mathbf{T}\boldsymbol{\beta})'\boldsymbol{V}^{-1}(\boldsymbol{x} - \mathbf{T}\boldsymbol{\beta}) \qquad (\cancel{4}3)$$

~~where~~Where **T** is ~~the~~ time~~,.~~, $\boldsymbol{x}$ is ~~the~~ observation data, and ~~and~~ $\boldsymbol{V}$ is the covariance matrix that can be estimated by Eq. (6) in Asmi et al. (2013). ~~The tFor technical details about the covariance matrix estimation refer to the description by Mudelsee (2010) and Asmi et al. (2013).~~

~~$\boldsymbol{\beta}$ = (parameter vetor),~~

~~$\boldsymbol{x}$ = (data vetor),~~

~~$\boldsymbol{T}$ = (time matrix),~~

~~and $\boldsymbol{V}$ is the covariance matrix. The estimated $\boldsymbol{V}$ matrix is:~~

~~= () × () × exp, (,=1,…,n) (5)~~

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~~, () are the variability of time series at t, t(). Here S is assumed to be time invariant, therefore is the stand deviation of the observation time series $x(i)$. ′ is the estimated, bias-corrected persistence time. To estimate the persistence time, the least-squares estimation is defined:~~

~~$S =$ (6)~~

~~and = argmin[S]. The minimization of S is done by Brent's search (Press et al., 1992).~~

~~After obtaining the covariance matrix $\boldsymbol{V}$, the solution of Eq.(6) is the GLS estimator:~~

~~$= \boldsymbol{T}'\boldsymbol{x}$ (7)~~According to the basic theory, the GLS fitting procedure is performed as follows:

~~(1) Approximate an initial estimation of parameters, $\boldsymbol{\beta}$.~~

~~(2) Based on the estimated $\boldsymbol{\beta}$,~~

~~Firstly, initial estimation of parameters $\boldsymbol{\beta}$ are approximated. According to the estimated $\boldsymbol{\beta}$, the trend,~~ and also seasonal and noise components ~~are obtained~~ estimated ~~from~~ from $x(i)$Eq. (2). ~~Then,~~ Moreover,lso tthe persistence time ′ and covariance matrix $\boldsymbol{V}$ ~~is~~are updated estimated from the residual.

~~(3) The covariance matrix $\boldsymbol{V}$ is used to estimate a new approximation of $\boldsymbol{\beta}$'.~~

~~(4) to iterate the GLS fitting unti~~Ifl the relative difference between the $\boldsymbol{\beta}$last two iterations of $\boldsymbol{\beta}$ from last two iterations is below a threshold 0.01 %, stop calculation. Otherwise, return to step (2) to iterate new estimation of $\boldsymbol{V}$.

To evaluate the robustness of the ~~estimated~~ derived slopes, the ~~the~~ ~~a~~Auto-~~r~~Regressive ~~b~~Bootstrap (ARB) method is~~was~~ ~~are~~ used to construct the ~~confidence intervals (~~CIs ~~) of the slopes~~ (Mudelsee, 2010, algorithm 3.5). ~~). Firstly, the residual $e(i)$ and persistence time are calculated from GLS approach. Then,~~The ARB resamples the white-noise residuals of data ~~by~~ using the auto-regressive persistence model ~~AR(1)~~, adds the resampled residuals to the fitted data, and re-calculates the slopes. The resampling ~~was~~ is repeated 1000 times and the CIs are~~were~~ estimated from these 1000 resampled slopes.

~~To ensure the comparability of trend slopes among different sites, the relative slope in % per year from both methods is used by dividing the absolute slope by the fitted median value of the first year~~whole observation time period~~.~~

2.3.3 Regional Mann-Kendall test

To detect if an overall increase or decrease exists in the whole region ~~studied~~, ~~R~~the regional Mann-Kendall test was also applied in this study. ~~The~~ Mann-Kendall test is a commonly used method for detection of ~~to detect the~~ long-term trends (Mann 1945; Kendall 1938). It detects the trend ~~by~~ using the Kendall's tau test~~,~~ which is known as a rank correlation test and ~~it~~ evaluates if a monotonic increasing or decreasing trend exists. If a significant monotonic increase or decrease is detected, the ~~a~~ Sen's slope estimator is further used to determine the slope and CI of the ~~corresponding~~ time series ~~based on Mann-Kendall test~~ (Gilbert, 1987). ~~To detect if an overall increase or decrease exists in a multi-site dataset, the regional Mann-Kendall test was extended to detect the trend over an observation network (Helsel and Frans, 2006).~~ The regional Mann-Kendall test (Helsel and Frans, 2006) ~~was~~is a method adapted from the seasonal Kendall test to determine whether a monotonic increase or decrease can be observed across a large area~~,~~ including multiple locations. ~~The technical~~ For details about the regional Mann-Kendall test ~~are given in~~refer to SM and ~~the article by~~ Helsel and Frans (2006).

For a time series $x(i)$ of length n, the ordinary Mann-Kendall statistic S is defined as

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~~$S =$ (8)~~

~~where~~

~~sgn = (9)~~

~~For large sample size (n>10), S is converted to a normal test statistic Z:~~

~~$Z =$ (10)~~

~~where the standard deviation of S is:~~

~~= (11)~~

~~A positive or negative Z refers to a monotonic increasing or decreasing trend. The significance of the trend can be evaluated by a two-tail test. At α = 0.05 significance level, the null hypothesis of no trend is rejected if >1.96.~~

~~Taking account of multi-sites, the regional Mann-Kendall test evaluates the individual Mann-Kendell statistic on each individual site k separately by Eq.(8), and sums of individual to obtain a regional Mann-Kendall statistic and then, ZL can be obtained:~~

~~= (12)~~

~~where the standard deviation of is:~~

~~= (13)~~

~~and is the number of the data at kth site.~~

~~Once the significant trend is detected, the slope can be evaluated by ordinary Sen's slope estimator (Sen, 1968). For a time series xi, the Sen's slope mL at each site L is:~~

~~= (14)~~

~~Then, the overall Sen's slope m is obtained by the median of those mL.~~ Considering the dataset size and calculation efficiency, the monthly median time series was used for the regional Mann-Kendall test in this study.

3 ~~Trends results~~Results and discussion ~~over the whole time period 2009‒2018~~

3.1 Overall trends over the time period 2009−2018

The temporal trends of the observed PNCs and eBC mass concentrations were evaluated using the customised Sen's estimator and GLS/ARB. ~~Table 3~~ The daily median time series ~~were~~ was used for the customized Sen's ~~estimator;~~estimator, and the monthly median time series ~~were~~was used for GLS/ARB. The relative annual slopes are listed in Table 3. ~~I~~The trend is marked as "statistically significant" (s.s.) in the table if ~~the observed trend~~it is statistically significant at 95 % confidence level~~, then the trend is marked as "statistically significant" (s.s.)~~. For the five parameters at the 16 sites (76 trends in total, see Table S1 in SM), the trends detected by the two methods agree well with each other with seven exceptions that we conclude as no s.s. trends~~, which we conclude as no s.s. trend~~ ($N_{[20–800]}$ at MEL, $N_{[10–30]}$ at LMI, LAN, HPB and SCH, and $N_{[200–800]}$ at LAN and BOS). The s~~S~~.s. negative slopes were found~~in~~ in 14 out of 16 sites for $N_{[30−200]}$ and $N_{[200−800]}$, in 11 out of 14 sites for $N_{[10−30]}$, and in all sites for eBC mass concentration and $N_{[200–800]}$. The annual slope of the eBC mass concentration varies between −13.1 % and −1.7 % per year. The slopes of the PNCs vary from −17.2 % to −1.7 %, −7.8 % to −1.1 %, and −11.1 % to −1.2 % per year (only the s.s trends) for 10−30 nm, 30−200 nm, and 200−800 nm particle diameter, respectively. ~~the sedby the two no clear trend for The annual slope of the eBC mass concentration varies between -13.1 % and -1.7 % per year. The slopes of the PNCs vary from -17.2 % to -1.7 %, -7.8 % to -1.1 %, and –11.1 % to -1.2 % per year (only s.s trends) for 10-30 nm, 30-200 nm, and 200-800 nm particle diameter, respectively. The r~~Robustness analysis ~~for the evaluated trends in the supplementary~~(see Sect. 1 in SM~~the SupplementSI.~~) suggests

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that the time span of our dataset is long enough for ~~true~~ slope detection ~~and that the influence of measurement uncertainty is negligible comparing with observed slopes~~. ~~The temporal trends of the PNCs and eBC mass concentrations were evaluated by the customized Sen's estimator and GLS/ARB. For the customized Sen's estimator, the daily median time series were used, while the monthly median time series for GLS/ARB. The relative annual slopes are shown in Table 3. Firstly, for 5 parameters at 16 sites (77 trends in total), two trend detection methods agree with each other very well, with six exceptions: $N_{[20-800]}$ at MST, $N_{[10-30]}$ at BOS, HPB and SCH, and $N_{[30-200]}$ at LAN and HPB, which we conclude as no increase or decrease. In general, significant decrease of the eBC mass concentration and $N_{[200-800]}$ are detected at all evaluated sites, except LAN, where no significant trends were found. The slopes of $N_{[10-30]}$ show high variability and lowest number of significant trends: at 7 sites there is a significant decrease and only MEL increase for both trend methods. Significant decrease of $N_{[30-200]}$ was found at all sites except LAN and three other regional background and mountain sites (MEL, NEU, HPB). In general, the annual slope of the eBC mass concentration varies between -7.7 % and -1.8 % per year, and the slope of the PNCs varies from -6.3 % to 2.7 %, -7.0 % to -2.0 %, and -9.5 % to -1.5 % per year (only significant trends) for 10-30 nm, 30-200 nm, and 200-800 nm, respectively. At site LAN, only insignificant decreases of the PNCs were detected. One speculation is that, due to its low data coverage at LAN, the trend detection methods might be hard to find the significant change. To detect if there is decrease of eBC mass concentration and PNC at LAN, we evaluated the trend of eBC mass concentration at another urban background site Raunheim. Site Raunheim is an urban background site of German Environment Agency (UBA), located in the city of Raunheim, about 15 km far away from LAN. The slopes of eBC mass concentration at Raunheim are -7.2 % and -5.9 % per year (both significant) for the customized Sen's estimator and GLS/ARB, respectively. It could be an indicator for reduction of eBC mass concentration at LAN.~~

~~On one hand, for diverse pollutant parameters and sites, their spatial representativeness is different due to the lifetime of pollutant and local influence (Sun et al., 2019). On the other hand, as shown in Table 3, not all the sites show the significant decreases of PNCs. Therefore, it is hard to conclude the regional reduction of the eBC mass concentration and PNCs all over Germany from the slopes evaluated at individual sites.~~ To evaluate the ~~regional variation of the~~overall trends of PNCs and eBC mass concentration ~~and PNCs all~~ over Germany, the regional Mann-Kendall ~~trend~~ test was ~~used~~applied to our dataset and the ~~regional trends~~ results are also ~~also shown~~listed in ~~the~~ Table 3~~is shown in Table 3 as well~~. It should be noted that roadside sites might bias the regional Mann-Kendall trends because of their prominent local influence.~~, three roadside sites might bias the result of regional Mann-Kendall test due to their prominent local influence.~~ Moreover, ~~the locations of~~ the ~~other~~ 13 non-roadside sites in GUAN are not evenly distributed in spatial scale ~~since~~ as there are ~~5~~ five sites located in the state of Saxony and HPB is only 42 km away from ZSF~~HPB and ZSF are only 42 km apart from each other~~. ~~This will result in a false trend throughout the entire region.~~ To ensure the representativeness of ~~spatial sampling~~the regional trends, three roadside sites (DDN, LMI and LEI) as well as LTR, ANA, and ZSF ~~are~~ were excluded in the regional Mann-Kendall test.

The highest regional decrease of 5 % ~~yr^{-1}~~ per year appears in the eBC mass concentration ~~of mass concentration~~ of which anthropogenic emissions are the major source. The regional trends of $N_{[30-200]}$ and $N_{[200-800]}$ ~~the PNCs in the size ranges 30–200 nm and 200–800 nm~~ are both s.s. negative. ~~$N_{[30-200]}$ represents the particles originated from anthropogenic emissions and the aged particles from new particle formation (NPF). Especially a~~In the~~A~~ urban area, $N_{[30-200]}$ and eBC mass concentration are found to be closely related to the emissions from incomplete diesel combustion (Cheng et al., 2013; Krecl et al., 2015). Therefore, the s.s. regional decreases in $N_{[30-200]}$ and eBC mass

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concentration ~~might~~are very likely to stem from the~~ indicate ~~ ~~declined~~decrease of anthropogenic emissions ~~is an important or even dominant driver for those decreases ~~in Germany. However, insignificant regional trend was detected for $N_{[10-30]}$~~10-30 nm~~. ~~One~~A plausible explanation ~~could be~~is that anthropogenic emissions have ~~probably~~ only minor or negligible influence on $N_{[10-30]}$ in the regional background area due to ~~their~~the short lifetime and high spatial variability of nucleation mode particles (Sun et al., 2019).~~The highest regional reduction rate appears on the eBC mass concentration of which anthropogenic emissions are the major source in Germany. The regional trends of the PNCs in the size ranges 30-200 nm and 200-800 nm are both significantly negative. $N_{[30-200]}$ represents the particles originated from anthropogenic emissions and the aged particles from new particle formation (NPF). Especially at urban area, $N_{[30-200]}$ and eBC mass concentration are found to be closely related to the emissions from incomplete diesel combustion (Cheng et al., 2013; Krecl et al., 2015). Significant regional decrease of $N_{[30-200]}$ and eBC mass concentration might indicate that, declined anthropogenic emission is an important or even dominant driver for those decreases in Germany. Insignificant regional trend was detected for 10-30 nm. One explanation could be anthropogenic emissions have probably only minor or negligible influence on $N_{[10-30]}$ at the regional background area due to the short lifetime and high spatial variability of young Aitken mode particles (Sun et al., 2019).~~

The trends of the PNCs and eBC mass concentration~~s~~ in this study are consistent with results from other studies conducted in Europe. Table 4 compares the long-term trends of aerosol ~~variables~~concentrations between the present and other studies~~ in Europe~~. Since 2001, the s.s. decrease in BC, PNCs, and $PM_{2.5}$ have been detected for most of the evaluated sites in ~~the t~~Table 4. The implementation of emission mitigation polices ~~were~~have been thought to be ~~one of ~~the dominant impact factors in these studies. Especially, there is one similar study that evaluated the trend of BC mass concentration in Germany~~,~~ for the time period 2005−2014 (Kutzner et al., 2018), in which~~. D~~decreased BC mass concentration was detected in 12 sites~~ in their study, due to the implementation of emission control legislation~~. Comparing the two studies, ~~One should be noticed that, the absolute slopes were presented in the study of Kutzner et al. (2018). T~~the absolute decreasing trend~~s~~ of BC mass ~~during~~for 2005-2014 is stronger than our results for 2009−2018, which might stem from the difference between the effects of emission mitigation policies in the two study periods.

~~. As the BC mass concentration continuously decreased since 2001, the mean concentration of BC mass during 2005−2014 is thought to be naturally higher than the one during 2009−2018. Thus, the stronger absolute decrease in BC mass during 2005−2014 does not represent the "faster" trend during 2005−2014 than 2009−2014.The trends of the PNC and eBC mass concentrations in this study are in consistent with studies in other European countries. In Europe, the negative trends of the total PNC, particle light absorption coefficient, and other optical properties were found at 9 regional background or remote sites from 2000 to 2010 (Asmi et al., 2013; Collaud Coen et al., 2013). In Spain, the PM_{10} and $PM_{2.5}$ decreased about -5.9 % and -6.0 % from 2004 to 2014, respectively (Pandolfi et al., 2016). The significant decrease of PM_{10} has been detected since 2008 due to the influence of reduced primary anthropogenic emissions in Po Valley, one large industrial manufacturing district in Europe (Bigi and Ghermandi, 2016). A similar study was conducted in UK. The BC trend from 2009 to 2016 varied between -0.62 % and -8 % at street, urban and rural background sites (Singh et al., 2018).~~

3.2 ~~Total e~~Emission change in Germany

Long-term trends of aerosol ~~variables~~concentrations on ~~the ~~regional scales could occur due to several factors such as emission changes and changes ~~of~~in long-range transport and vertical diffusion due to inter-annual variations of

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weather and climate ~~change which result in long-range transport and vertical diffusion~~. In areas strongly affected by ~~strong~~ human activities (e.g. traffic, domestic heating, industry etc.), changes in emissions are ~~generally~~usually the main cause of the trend of aerosol concentrations~~(Kutzner et al., 2018; Bigi and Ghermandi, 2014)~~. From 2005, the German Federal Government has ~~set the goal of~~established ~~the~~ German Sustainability Strategy with ~~a~~the goal of ~~to~~ reducing~~e~~ the emissions of SO_2, NO_x, NH_3, non-methane volatile organic compounds (NMVOC), and $PM_{2.5}$. ~~Emission of all primary and precursors decreased from 2000 to 2016 in the European countries. PM2.5 and BC emission decreased about 20 % and 40 % between 2000 and 2016 (European Environmental Agency, 2018). Therefore, the trend obtained in the previous section is most likely caused by emission reduction. In this section the reason for decreasing trends will be explored based on emissions change in the time period of 2009-2017.~~ Figure 3a illustrates the variation of BC total emission ~~of BC~~ in Germany from 2009 to 2017 (black line) and the annual mean eBC mass concentration index (defined as the percentage of the ~~mean~~ concentration for~~in~~ the year of 2009) for the six regional background and mountain sites (magenta line)~~, defined as the percentage of the mean concentration in the year of 2009 (% of 2009)~~. From 2009 to 2017, the total emission of BC in Germany decease~~d~~s about −3.4 % per year and ~~it shows~~ highly agree~~ments~~ with the ~~change~~trend of mean eBC mass concentration. ~~As a tracer of anthropogenic emission, the~~ BC mass concentration is ~~controlled~~influenced by ~~the~~ emission, transport, and scavenging simultaneously. ~~Highly agreed change~~The agreement between the ~~of~~ trend of BC total emission and eBC mass concentration suggests that emission reduction is very likely to be the ~~most~~ dominant ~~impact~~ factor for the ~~reducedin~~decreasing of ~~-~~eBC mass concentration ~~in~~over Germany, while ~~the meteorological condition and long-range transport~~ other factors does not show a clear contribution.

~~Different with BC, b~~Other than primary emission~~the emitted primary aerosol particles~~, another important process controlling the PNC is the formation of secondary ~~particles~~particulate matters. ~~The relative secondary organic aerosols (SOA) contribution was higher in rural background condition than in urban sites (Castro et al., 1999).~~ The decreased anthropogenic emissions might ~~may also~~ reduce the concentration of precursor gases and thus inhibit secondary aerosol formation~~secondary particles by limiting the emission of precursor gases~~. Figure 3b illustrates the ~~emission trend~~variation of total emission of $PM_{2.5}$ and some selected precursor gasses, as well as the annual ~~change of~~ mean PNCs index for the six regional background and mountain sites. It should be noted that ~~The~~ PNC is not a conserved parameter ~~,~~ and ~~it~~ might change rapidly ~~during~~by particle coagulation~~, but particle volume concentration (PVC) and particle mass do not~~. Thus, another parameter, ~~total~~ particle volume concentration (PVC) in the size range of 20−800 nm ($V_{[200-800]}$), is also ~~evaluated~~shown ~~-~~in ~~the~~ Fig. 3b ~~, ensuring the~~for a better comparability ~~between sub-micrometer aerosol particles and PM2.5 emission~~. Total emissions of all precursors and $PM_{2.5}$ except NH_3 decreased around −2.2~−0.9 % per year during 2009−2018~~,~~. However, the measured ~~slope of~~ $N_{[30-200]}$, $N_{[200-800]}$ and $V_{[200-800]}$ decreases about −2.5 % per year, which is stronger than the decreases in ~~PM2.5 and precursors~~ the emissions ~~(-0.9~2.2 % per year)~~. This ~~may~~might be because the secondary aerosol contributes a large fraction in particulate matters in the regional background settings (Castro et al., 1999). Secondary aerosol formation processes are highly complex and nonlinear, determined ~~by~~ not only by the concentrations of precursors but also many other factors such as solar radiation, temperature, humidity, and diffusion conditions etc. The change ~~of~~in secondary aerosol particle concentration ~~may~~might not follow the change of precursor emission. Therefore, larger discrepancies are observed between the emissions and particle concentrations although decrease trends are found in both of them.~~There are two plausible explanations: First, the secondary particles contribute larger mass fraction in the regional background setting than urban area (Castro et al., 1999). The process of secondary particles formation is complex, and its reaction paths are numerous and highly~~

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~~nonlinear. So, the change of secondary aerosol particle concentration may not be linear with the change of precursor emission. Moreover, the formation of secondary aerosol particles is controlled by a number of factors, including precursor gasses, solar radiation, temperature, humidity and diffusion conditions etc. These factors will be affected by weather condition. In previous discussion of BC emission reduction, we concluded the weather condition have only negligible impact on the decreased eBC mass concentration. However, weather condition may have a greater effect on secondary aerosol particles than on BC, resulting in the different trend of~~ ~~$PM_{2.5}$~~ ~~and precursors emission and trend of PNC. The reduction of~~ ~~$N_{[10-30]}$~~ ~~is stronger than PNCs in other size ranges in Fig. 3b.~~ ~~$N_{[10-30]}$~~ ~~mainly represents newly formed particles, which has spatial inhomogeneity in most of the cases (Sun et al., 2019). Therefore, the representativeness of~~ ~~$N_{[10-30]}$~~ ~~might not be as good as other parameters, since the uncertainty is relatively high.~~

Based on the above results, we believe that the observed trends of PNCs and eBC mass concentration are ~~is~~ mainly due to the reduction in emissions. The annual changes of meteorological conditions might have an impact on PNCs, but are not likely to be the decisive impact factor. Detailed discussion on the possible influence of meteorological conditions will be discussed in Sect. 4. The decreased pollutant concentrations are highly associated with the reduced risk of human health. Pope et al. (2009) demonstrated that a decrease of 10 µg m^{-3} in the $PM_{2.5}$ mass concentration is related with an increase of life expectancy of 0.61 ± 0.20 year in 211 counties. The improved health effects because of decreased UFP and BC would be even greater compared with that of $PM_{2.5}$ mass concentration. As of 2018, 97 % of cities in low- and mid-income countries do not meet the World Health Organization (WHO) air quality guidelines (WHO, 2018). Our result demonstrates that the implementation of proper emission mitigation policies can largely reduce the BC mass concentration and PNC, thus may effectively reduce the health risk in polluted regions.

~~The annual changes of weather condition might have an impact on PNCs, especially on~~ ~~$N_{[10-30]}$~~~~, but will not be the decisive impact factor. The influence of weather conditions will be discussed in detail in Sect. 4.~~

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~~3.2 Robustness of the trends~~

~~For the time series of a climate parameter, its trend may be caused by the homogenous variations in meteorological conditions or aerosol emissions (Conrad and Pollak, 1950), but sometimes also can be caused by inhomogeneous "break points" such as site relocation, inlet change, and new pollution sources (Collaud Coen et al., 2013). The break points not only make the time series inhomogeneous but also result in a poor representativeness of the trend. Normally, only the trends of homogenous time series are considered to be robust and trustable. Another important factor affecting the trend is the size of the time series. As shown in Fig. 2, the sizes of the time series are not the same for all evaluated sites, vary from 6 to 10 years. To evaluate if the detected decreases or increases are homogeneous and if our dataset is long enough to provide the robust trend, the evolution of trend was analyzed. Fig. 3 shows the annual changes of the eBC mass concentration and PNCs for expanding time intervals starting from 2009, using the customized Sen's estimator. The average trend for each site category is illustrated. It can be seen that, the trends tend to be stable without strong variation after time interval 2009-2016, indicating our dataset is sufficient for true slopes.~~

~~Gaps in time series may bias the observed trends. Generally, it is difficult to quantify clearly the influence of data gaps on the trend results. In this study, since the influences of periodicity and outliers are diminished by the customized Sen's estimator, the evaluated trends are less sensitive to data gaps than those derived by other methods. Still, data gaps may affect the trend results especially for sub-dataset, for example the trends in particular seasons.~~

~~4 Trend in sub-sets~~

~~As shown in Sect. 3, declined anthropogenic emissions are very likely to be the main factor of the decreased PNCs and eBC mass concentration in Germany. The intensity of human activities such as traffic volume usually has weekly and diurnal cycles. To further investigate the role of anthropogenic emissions in the downward trend of the PNCs and eBC mass concentration, their weekly, diurnal and seasonal trends were analyzed in this section.~~

~~4.1 Weekly trends~~

~~For the weekly Sen's slope, only the data pairs belonging to the same weekday were selected to calculate the slope *m*. Figure 4 illustrates the average Sen's slopes of the PNCs and eBC mass concentration for working day (from Monday to Friday) and weekend (Saturday and Sunday) at each site category.~~

~~At roadside where traffic emission dominates the PNCs and eBC mass concentration, higher reduction rates are observed on working days for all five parameters. Traffic emission has direct influence on urban background aerosol, thus reduction rates at urban background sites are higher on weekday. But the differences are smaller than those for roadside. This result implies that traffic emission control policies such as LEZ is a main factor of the decreases of the PNCs and eBC mass concentration in urban area. There is no significant difference can be seen between working day and weekend for the regional background, low mountain range and high Alpine sites, rather indicating that the cause for the decrease is far away from the background condition and hence closer to urban areas.~~

43.~~2~~ 3 Diurnal variation of trends

The ~~intensity of~~ emission intensit~~y~~ies ~~from~~of some emission sources have distinct ~~seasonal~~diurnal variations, such as that of traffic ~~(e.g. morning and evening rush-hours)~~ and residential activities (e.g. domestic heating). T~~,T~~he trends based on the subsets of ~~data~~the time series might reflect the impact of these changes. In ~~this and the following section~~this section we will analyse the diurnal ~~and seasonal~~ variations of trends and investigate their connection to the sources.

Figure 4 shows the ~~customised~~ Sen's slopes of the measured PNCs and eBC mass concentration at each hour of day for each site category. To evaluate the diurnal trends, data pairs belonging to ~~a particular~~the same hour of day were selected ~~to calculate the slope *m*~~for the calculation of Sen's slope. ~~in~~ Figure 4 shows~~ure shows~~ that all the four parameters show distinct diurnal variations at the roadside and urban background sites ~~all four parameters show distinct diurnal variation at roadside and urban background sites~~. ~~In~~At the roadside sites, the decrease in eBC mass concentration, $N_{[10–30]}$ and $N_{[30–200]}$ are much stronger during daytime ~~the~~ than ~~in~~during ~~night-time~~night-time ~~than in the daytime~~. ~~In~~At the urban background sites, the diurnal trends ~~of eBC mass concentration and PNCs~~ also show stronger decrease in the morning and ~~also in the~~ evening~~, although the decrease is not as strong as in the roadside sites~~. Such ~~a~~ diurnal patterns ~~of trend is~~are consistent with the diurnal variation of ~~motor~~ vehicle volume ~~variation~~ in the ~~city~~urban area~~. In urban areas,~~ where traffic emission is ~~thought to be~~ the dominant source of BC and ~~ultrafine particulate matter~~UFP (Ma and Birmili, 2015). As shown in Fig. S2 in SM~~the Supplement~~~~in~~, road transport ~~emission~~ contributes to the highest ~~emission~~ reduction ~~s~~ in total emission of ~~the emission of~~ BC, $PM_{2.5}$, and precursors NO_x and NMVOC, from 2009 to 2017. Therefore, it can be concluded that the daytime strong~~er~~ ~~decrease~~reduction ~~of~~in PNCs and eBC mass concentration in the ~~the~~ urban area ~~during the daytime~~ is a ~~direct~~ result of the ~~reduction~~decrease ~~in~~in~~of~~ road transport emissions. In Germany, the government has made great effort to reduce the emissions due to road transport. The 10th BimSchV (first~~ly~~ issued in 1994 and entered into force of

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recast on 14 December~~ 14~~, 2010) regulates the emission requirement for petrol, diesel, and bio-diesel. ~~The regulation firstly issued in 1994, and then entered into force of the recast on December 14, 2010. The 20th BImSchVregulates the emission limit of volatile organic compounds (VOC) during transport and storage of petrol and fuel mixtures. This regulation limits the emission of precursors, further reduces the PNC near traffic condition. Another regulation (~~And the 28th BImSchV (issued in 2014 and amended every year)~~)~~ regulates the type of engines for mobile machinery that can be marketed commercially, ensuring low emissions from new commercial vehicles. Meanwhile, the implementation of the European Emission Standard (EURO standards, https://en.wikipedia.org/wiki/European_emission_standards, last access: 18 September~~ 19~~, 2019) starting from 1990s has significantly reduced the emissions from gasoline and diesel engines. ~~It defines the limits for exhaust emission of new vehicles sold in EU and EEA member states (https://en.wikipedia.org/wiki/European_emission_standards, last access: September 19, 2019) and it is now widely used by car manufacturers.~~ Moreover, another regulation LEZ (35th BImSchV) has ~~can~~ effectively reduced the traffic emissions by restricting ~~the~~ highly polluting vehicles in defined area in the city. Resulting from the above ~~combination effect of those~~ policies ~~above~~, the road transport emissions of BC, $PM_{2.5}$, NO_x, and NMVOC have significantly decreased during 2009−2017 as shown in Fig. S2. ~~ThiAnd~~ O~~o~~ur results~~s~~ ~~result~~ confirm~~s~~ that the reduction in traffic emissions plays a main role in the decreasing trends of eBC mass concentration in Germany, especially in the urban area.~~Figure 5 shows the customized Sen's slopes of the PNCs and eBC mass concentration at each hour of day. Similar to the weekly trend, data pairs belonging to a particular hour of day were selected to calculate the slope *m*.~~

~~For BC which is mainly emitted from anthropogenic sources in Europe, diurnal patterns with higher reduction rate in daytime than in night-time can be seen at roadside sites. Reduction of traffic emission can directly cause a decrease of eBC mass concentration in near source areas. Therefore, higher reduction rate is observed in daytime when human activities are more intensive. Negative slopes can be also observed in night time and in other site categories. A plausible explanation is that, reduction of local anthropogenic emissions can also reduce the background eBC mass concentration in a larger area and longer time scale since BC has a lifetime of around a week (Cape et al., 2012; Wang et al., 2014). This result confirms that reduction of anthropogenic emissions plays a main role in the decreasing trends of eBC mass concentration in Germany.~~

~~The trends of the PNCs depend on the particle size ranges and time of day. In most of roadside sites, similar diurnal patters as that for eBC with higher reduction rate in daytime and lower rate in nighttime can be observed for $N_{[20\ 800]}$, $N_{[10\ 30]}$ and $N_{[30\ 200]}$. In cities, traffic emission may have large contribution on PNC in these size ranges, thus we attribute this diurnal pattern of reduction rate also to the reduced traffic emission in urban background conditions, similar as to eBC mass. NPF is an important natural source of ultrafine particles and may largely enhance $N_{[10\ 30]}$. Based on the GUAN dataset, Ma and Birmili (2015) reported that the annual average contributions of NPF on $N_{[5\ 20]}$ are 12 %, 24 % and 54 % at roadside, urban background and regional background sites, respectively. Therefore, the inter-annual change of NPF frequency or intensity may also determine the trend of $N_{[10\ 30]}$ especially in urban and regional background sites. Actually, as can be seen in Fig. 5c that $N_{[10\ 30]}$ show a maximum reduction rate of around -3 % in the afternoon at the regional background sites. It is likely to be resulted from the inter-annual change of regional NPF events since NPF is the only dominant source at those sites. At regional and mountain sites, $N_{[30\ 200]}$ and $N_{[200\ 800]}$ show a constant negative trend throughout the day, suggesting the decrease of PNCs in the regional background air which is likely to be the result of the reduction of anthropogenic emissions in cities.~~

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For regional background and low mountain range sites~~,~~ that are ~~generally~~ far from the road ~~the~~ traffic and ~~will~~ not ~~be~~ directly affected by traffic emissions, trends of eBC mass concentration and PNCs do not show ~~the~~ distinct diurnal patterns~~,~~. ~~but the~~ ~~But~~However, downward trends ~~can be still seen~~are still visible, which stems ~~from~~ mainly from the decrease ~~of~~in background concentration in the whole region caused by the reduction ~~of~~in emissions as shown in Fig. ~~5~~S2. ~~Especially for eBC mass concentration, the decrease is more obvious. As discussed in the previous section, the dominant factor for the decreasing trend is the decrease of background concentration in the whole region caused by the reduction of traffic emissions. For PNCs, the decrease of other emission sources is also crucial, such as decreased emission of $PM_{2.5}$, SO_2, NO_x and NMVOC on the sector of residential, other fuel combustion or industry (see Fig. 5). However, as shown in Fig. 5, one of the most important contributors for the decreasing trend is still the decrease of road transport emission. When there are fewer motor vehicles at night, the PNC in the city sites also show a significant decline, which is also thought to be due to the background concentration decrease caused by the reduction of other emissions mentioned above.~~ For the low mountain range and high Alpine sites, trends of eBC mass concentration, ~~$N_{[10-30]}$ and~~ $N_{[30-200]}$ and $N_{[200-800]}$ ~~also~~ show weakly ~~decrease on~~ diurnal ~~trend~~patterns with slightly ~~higher decrease~~more reduction in the afternoon~~, only slightly decease can be seen in the afternoon~~. This is mainly ~~related to the station height. In the afternoon,~~because the high-altitude sites ~~has~~might~~ay~~ have more chance to ~~stay~~merge into the planetary boundary layer (PBL) in the afternoon~~due to evolution of PBL~~, resulting in a much stronger influence of anthropogenic emission~~s~~decrease.

It is interesting ~~to notice~~ that at the urban sites $N_{[10-30]}$ has similar diurnal pattern as other parameters, but at regional background and low mountain range sites they look quite different. ~~For~~At the regional background sites, $N_{[10-30]}$ shows a maximum average reduction rate of around ~~−3~~1.5 % per year ~~%~~ in the afternoon ~~at the regional background sites~~but basically zero trend during the night~~(see Fig. 2 in the supplementary)~~. New particle formation ~~(should be noting that the diurnal trend of $N_{[10-30]}$ is similar to other parameters for the roadside site category, but they are quite different with the regional background and low mountain range sites. In fact,~~ NPF) is a ~~important~~dominant source of $N_{[10-30]}$ in the non-urban areas. ~~Based on the GUAN dataset,~~ Ma and Birmili (2015) reported that the annual average contributions of NPF on $N_{[5-20]}$ ~~are~~is ~~12 %, 24 % and~~ 54 % at ~~roadside, urban background and~~ regional background sites~~, respectively~~. ~~The influence of NPF needs to be considered when we analyse the trend of $N_{[10-30]}$. Most of the NPF events occur in the daytime with strong sunlight. So, t~~Also~~nd~~ the ~~long-term trend resulting from~~ contribution of NPF ~~events can~~has~~ight also show~~ a diurnal ~~trend~~pattern with higher levels in the afternoon and no influence during~~at~~ night. Thus, ~~For regional background sites, $N_{[10-30]}$ show a maximum reduction rate of around -3 % in the afternoon at the regional background sites (see Fig. 2 in the supplementary). It~~the diurnal variations in~~of~~ $N_{[10-30]}$ trend in the~~at~~ regional background sites is likely to have resulted from the inter-annual changes in the regional NPF events~~since NPF is the only dominant source at those sites~~. ~~In the night-time, there is no trend can be seen for $N_{[10-30]}$ at regional background sites, due to the absence of source for UFP. For roadside sites, traffic emissions contributed more than 50 % of $N_{[10-30]}$ (Ma and Birmili, 2015). Thus, stronger decrease can be seen during daytime, similar to other parameters. For urban background sites, the contribution of NPF accounts for about 50 % (Ma and Birmili, 2015). Therefore, besides the traffic emission, the NPF may also contribute to the decrease in the daytime. There is no obvious pattern a~~At the low mountain range sites, statistically insignificant ~~but the~~positive trends are observed~~ positive~~. One possible reason is that the low mountain range sites are far from emission sources of nucleation mode particles (e.g. traffic) and NPF is also rare in the areas. Thus the trend of $N_{[10-30]}$ might be more influenced by metrological conditions.

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~~theHerrmann et al., 2015,ightAs discussed in the Sect. 3.2, the reaction path and influencing factors involved in NPF are very complex. In addition, the life time of nucleation model particles is relatively short, and they may grow to the accumulation mode in a relatively short time, leading to the high variability of $N_{[10\text{-}30]}$ on the spatial scale. Therefore, even if the precursors emission such as SO_2 reduced, positive trends can still be observed in areas such as low mountain range area.~~

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~~4.3~~3.4 Seasonal variation of trends

Figure 5 shows the ~~statistical results of the multi-annual~~ trends of the PNCs and eBC mass concentration~~s~~ for ~~different~~ each seasons. In general, negative trends are found for the five parameters ~~in~~ in ~~most of~~ all seasons ~~and pollutant parameters~~. Similar seasonal trend patterns with stronger decrease rate~~s~~ in winter are detected for all PNCs~~, but not for eBC mass concentration. Stronger decrease of PNC in winter~~ which is ~~very~~ likely to have been caused by ~~some other~~ factors that have strong seasonal variation~~, for example the~~ such as domestic heating and/~~combustion,~~ or ~~the inter-annual changes in weather~~meteorological conditions. The emission of ~~The emission of d~~domestic heating is much stronger ~~a rate occurs usually in winter~~during~~in~~ cold season. The 1st BImSchV limited the emission for medium ~~or~~and small combustions (e.g. domestic heating). ~~As can be seen from Fig. 5,~~ Although domestic heating (~~the~~ residential sector) contributes only a minor fraction of~~in~~ the total emission ~~the emission~~ of BC, $PM_{2.5}$, SO_2 and NMVOC, ~~in the residential sector decreased during 2009-2017,~~its absolute decrease from 2009 to 2017 ~~in~~is large and comparable with other sectors (Fig. S2~~1~~) ~~although they contribute only minor mass fraction in the total emission~~. ~~Conversely, t~~The least decreases rates in~~of~~ the PNCs were found in summer. ~~Besides~~Other than the ~~residential emission is~~ low residential emission in ~~summer~~warm seasons, another ~~impact factor~~ reason ~~might~~mightay be the strong seasonal variations in~~of~~ biogenic emissions (Asmi et al., 2013). Biogenic emissions contribute considerable secondary organic aerosol (SOA) precursors in summer and thus a higher ~~ighthave s~~contribution on PNCs~~trong seasonal variation with higher emissions in summer~~. Without any strong ~~long term~~long-term variation, ~~T~~the stable contribution of ~~higher~~ biogenic emissions on PNCs ~~in summer will~~mightight ~~mask~~lower the relative decrease rates in~~of~~ PNCs ~~caused by anthropogenic emissions~~duringinin summer. ~~However, n~~No clear seasonal pattern could be observed for eBC mass concentration because its emission decrease is mostly contributed by the road transport that~~which~~ has no obvious seasonal variation. ~~the sector which contributes the largest mass fraction and the strongest decrease is not the residential, but the road transport which has no obvious seasonal variation.~~

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~~Conversely, the least decreases of the PNCs were found in summer. Besides the residential emission is low in summer, another impact factor might be the seasonal variation of biogenic emission (Asmi et al., 2013). Biogenic emissions contribute considerable SOA precursors and have strong seasonal variation with higher emissions in summer. The higher biogenic emission in summer will mask the decrease caused by anthropogenic emission.~~ Long-term change of meteorological parameters might also affect the seasonal trends as well~~. It~~ and will be discussed in ~~the~~ Sect. 4.~~It is obvious that the seasonal change of weather condition will have an influence on the change of PNCs and eBC mass concentration. In the warm season, the higher plenary boundary layer (PBL) height and better dilution can reduce the PNCs and eBC mass concentration, but NPF events may increase the PNC especially the nucleation mode particles~~ ~~$N_{[10\text{-}30]}$. Conversely, PNC and eBC mass concentrations are elevated in cold season due to a less mixed PBL and higher anthropogenic emissions such as domestic heating. In this section, the seasonal trends of the eBC mass concentration and PNCs were detected. For seasonal trends, only the data pairs belonging to a particular season were used to calculate the customized Sen's slope~~ ~~m.~~

3.5 ~~The influence of l~~Evaluation of low emission zones

As discussed in the previous sections, the reduction in total emissions could be ~~directly~~ reflected directly in the long-term trends of the aerosol observation ~~network~~ in Germany, suggesting~~y. This also shows~~ that long-term observation network with different types of site is an effective tool to verify the effectiveness of emission control policies~~ reductions~~. However, the observed decreas~~ing~~e in trends ~~we observed is~~is ~~aare~~ ~~most likely to be the combination~~ combined result of the various emission mitigation policies. A question raised is that can such long-terms observation network reflect the effect of a specific emission mitigation policy. ~~Therefore, can such an observation network dataset effectively reflect the effectiveness of one single specific emission mitigation policy?~~ LEZ is believed to be a good candidate for such an evaluation due to several reasons. Firstly, LEZ has usually a clear introduced date in each city. Secondly, traffic emissions are~~is~~ the major source of aerosol particles in the urban area. Thirdly, traffic emission sources are basically evenly distributed in the urban area and therefore,~~hus~~ its contribution of~~n~~ particulate matter in the urban area will not be strongly influenced by wind direction. ~~because the traffic emission is the major source for the BC and aerosol particles in the city and the LEZ has clear introduced date in each city. Second, the traffic emission source is not a point source which means it cannot be strongly influenced by wind speed, wind direction or other impactors. Third, LEZ is a policy that the high polluting vehicles are prohibited to enter and is easy to be reflected in the observation measurement.~~ In this section, we will analyse the effects of ~~the~~ two LEZs ~~on the aerosol concentrations~~ in the urban area based on our dataset.

LEZ is an urban access regulation in Europe~~, in which, the most polluting vehicles are regulated. LEZ~~ and is one of the key ways to reduce traffic emissions in urban areas. In Germany, high-, medium-, and low-emitting vehicles are required to be marked with ~~with~~ red, yellow or green colour stickers on the front window shield. ~~a particular sticker with red, yellow or green color is required on the car, banning the high-, medium- and low-emitting vehicles.~~ The green sticker denotes the diesel vehicles with at least Euro 4 or Euro 3 engines with a particular filter and petrol vehicles meeting ~~with~~ at least Euro 1 standard. Vehicles with green stickers have lowest emissions and can enter all LEZs. Vehicles with other ~~sticker or~~ stickers, meaning higher emissions, are restricted. ~~The yellow one indicates the diesel vehicle with at least Euro 3 or Euro 2 with particular filter; the red one is for the diesel vehicles with Euro 2 or Euro 1 with particular filter. Different sticker indicates the different area allowed to enter. The diesel vehicles with Euro 1 or worse limit, or the petrol car without a catalytic converter have no sticker, which means that, they are not allowed to enter LEZ.~~ In Germany, the first LEZ was launched ~~introduced~~ in 2008 in Berlin. As of November 2019, LEZs are implemented in over 60 cities. ~~The s~~Short-term studies ~~showed~~have shown that~~,~~ LEZ can immediately reduce the pollutant levels after the implementation ~~, resulting from absence of highly polluted vehicles in LEZ~~ (Rasch et al., 2013; Qadir et al., 2013; Jones et al., 2012).

We select two cities that have implemented LEZ during 2009−2018 and have measurements of both PNCs and eBC mass concentration ~~and PNC measured~~: Leipzig and Augsburg. Figure 6 illustrates the deseasonalised ~~monthly~~time series of monthly averaged parameters measured ~~for~~at the two urban background sites AUG and LTR, by subtracting the seasonal cycle from the mean monthly time series. ~~And t~~The horizontal dashed lines in Fig. 6 denote the mean values with respect to different LEZ stages. In Augsburg, the first, second, and third stages of the LEZ were implemented respectively ~~came into force~~ on 1 July, 2009 (red dashed line), 1 ~~the second stage on~~ January, 2011 (yellow dashed line), ~~the third stage on~~and 1 June, 2016 (green dashed line). ~~Figure 7 (c) and (d) show the deseasonalized eBC mass concentration and $N_{[30-200]}$ (gray line) and mean value of gray line with respect~~

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~~to the different LEZ stages.It can be seen I~~iFigure 6a and b show that the~~,~~ ~~the concentration of~~ eBC mass concentration and $N_{[30–200]}$ have gradually decreased after the implementation of ~~the~~ each ~~starts~~ of new stages of LEZ ~~introduced, due to the absence of high polluting vehicles~~. However, the difference between stage 2 and 3 is relatively negligible ~~in AUG~~. A possible reason is that ~~The reason is,~~ the third stage of LEZ came into force in June 2016 in Augsburg. By June 2016, around 52 cities in Germany had already implemented the third stage of LEZ,~~,~~ which accelerates the fleet update in the whole country (also in Augsburg). ~~A large number of vehicles which did not meet the requirement of the third level, have already completed the fleet turnover.~~ Thus, no large difference could be seen between the second and third stage of LEZ in the city of Augsburg. It is worth noticing that even with the seasonal variation subtracted from the time series, the amplitude of short-term variations of eBC mass concentration and $N_{[30–200]}$ are still very large mostly due to variations in~~of~~ meteorological~~y~~ conditions. Sometimes it is even larger than the concentration decrease caused by the implementation of LEZ. This indicates~~means~~ that short-term measurement might be influenced largely by the variations in meteorological conditions and long-term measurements are necessary for a trustworthy verification of~~such as~~~~like~~ LEZs.

The LEZ was entered into force in the city of Leipzig directly on third level on 1 March, 2011~~, directly on third level.~~ ~~Therefore,~~ Clear decrease is observed in~~the~~ $N_{[30–200]}$ ~~eBC mass concentration and~~ ~~$N_{[30–200]}$~~ ~~show clear decrease~~ after 2010 (~~shown~~Fig. ~~7~~6 ~~((~~c) but nearly invisible in eBC mass concentration (Fig. 6d)~~)~~. In ~~the city of~~ Leipzig, ~~we have measured~~ the PNCs and eBC mass concentrations ~~and PN~~were measured ~~C~~ at both roadside and urban background sites simultaneously, which provide us the possibility to directly detect the traffic contribution by evaluating the ~~traffic~~ increment of the aerosol concentration (the difference of the concentrations between the traffic and the background sites). ~~And t~~The effects of background variation, other sources and meteorolog~~y~~ical factors can be ignored. Figure 7 illustrates the annually averaged diurnal cycles of the ~~traffic~~ increment ~~for measured parameterss~~. Before and after the implementation of LEZ ~~implementation~~ (2010 and 2011), the PNCs and eBC mass concentration show a sudden decrease of up to 40 % during daytime~~, especially,~~. ~~the eBC mass concentration decreased about 50 %.~~ The mean concentration of eBC mass, $N_{[30–200]}$ and $N_{[200–800]}$ during working hours (06:00 to 18:00 local time) in ~~the year of~~ 2010 are respectively about 1.63, 1.33, and 1.58 times higher than ~~the year of~~ those in 2011. After 2011~~the year of 2011~~, these aerosol variables continued to decrease, ~~which is~~ mainly due to the continuous~~ly~~ update of vehicle fleet ~~until 2018~~. ~~Actually,~~ These~~isis~~ result suggests that with a multiple-site network, the effect of emission control policy ~~can~~could be better detected from the increments between near-source and background sites.

~~the effects of other sources or other factors can be ignored from the traffic increment. The decrease after 2011 may have been due to the slow replacement of some high polluting vehicles. It confirmed that, the LEZ can effectively reduce the traffic emission as well as traffic-related pollutants in the urban area. It also shows that our long-term observational dataset can be used as a good indicator of the effectiveness of such emission mitigation policy like LEZ.Figure 6 shows the statistical results of the multi-annual trends of the PNC and eBC mass concentrations for different seasons. In general, negative trends are found in all sites and pollutant parameters except~~ ~~$N_{[10-30]}$~~~~. Reductions of the PNC are found to be stronger in winter, which can be regarded as a result of the implementation of the emission mitigation regulations for large or small combustions, such as domestic heating or power generation in winter. Conversely, the least decreases of the PNCs were found in summer. One impact factor might be the seasonal variation of biogenic emission (Asmi et al., 2013). The biogenic emission increases in summer, which will mask the decrease caused by anthropogenic emission. In winter, less biogenic emission makes anthropogenic emissions more prominent. Therefore, a higher decrease can be seen in winter, indicating that the~~

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~~decreasing trends of the PNCs are more likely related to anthropogenic sources than biogenic ones. Long-term change of meteorological parameters may affect the seasonal trend as well. It will be discussed in the next section.~~

~~5~~ 4 Meteorological influences on the trends of ~~the~~ particle number and ~~the~~ eBC mass concentration

Meteorological conditions also influence the concentration of ~~temporal variation of~~ aerosol particles (Birmili et al., 2001; ~~; Mikkonen et al., 2011;~~ Spindler et al., 2013; von Bismarck-Osten et al., 2013; ~~Wehner and Wiedensohler, 2003;~~ Hussein et al., 2006) and~~.~~ their~~Thus its~~ ~~L~~inter-annual~~ong-term~~ changes~~s of meteorological conditions (precipitation, PBL height, wind speed, temperature etc.) could~~ might~~ay~~ ~~cause~~modify the trends of the parameters studied ~~increase or decrease of atmospheric pollutant concentration.~~ ~~.~~In this section, t~~To investigate t~~he potential ~~contribution of~~influence of ~~possible changes in~~ meteorological conditions, including ~~in the period of interest, trends under different weather conditions~~ precipitation, temperature, wind speed, and air mass types~~,~~ are discussed ~~in this section~~.

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~~5~~4.1 ~~Seasonal trends of~~Influence of precipitation, temperature, wind speed on the detected trends ~~meteorological parameters~~

Table ~~4~~ 5 provides the long-term trends of precipitation, ambient temperature, and wind speed ~~all over Germany for the period~~during 2009–2018 based on the 76 measuring sites distributed all over Germany. The trends of the parameters were evaluated by the customised Sen's estimator. The ~~meteorological~~ data was obtained from Germany's National Meteorological Service (Deutscher Wetterdienst, DWD). ~~The daily values of these three meteorological parameters at 76 measuring sites in Germany were provided. All~~The 76 DWD sites are grouped into three categories: urban background, regional background, and mountain area. ~~The mean time series among all 76 sites~~sites in each category ~~were used as the area average of meteorological data in Germany. The trends of meteorological parameters were evaluated by the customiszed Sen's estimator~~T~~It can be seen that t~~he trends~~s~~ in~~of~~ all three meteorological parameters agree well among the three categories~~regional~~. Temperature shows a negligible change in spring, a slight increase in summer~~,~~, and a larger increase up to~~reduction~~ 0.43 ℃ per year~~yr-1 only~~ in autumn and winter. Increased temperature ~~, especially~~ during~~in winter~~ cold seasons ~~,~~might have~~ay lead~~led to lower anthropogenic emissions from domestic heating ~~or~~and power generation, ~~thus may contribute on~~resulting ~~in~~and further led to a decrease in~~of~~ PNCs and eBC mass concentrations~~5~~6. Precipitation presents a s.s. decreas~~ing~~e trend up to about −6 % per year~~yr-1~~ in summer, which might~~ay~~ inhibit the wet deposition of aerosol particles and diminish the reduction of eBC mass concentrations and $N_{[200-800]}$ to a~~some~~ certain extent. No obvious trend is observed in wind speed. In summary, increased ambient temperature might contribute to the decrease of the PNCs and eBC mass concentrations shown in Sect. 3.1 and 3.4 by indirectly influencing anthropogenic emissions. While decreased precipitation in summer might diminish the decrease of the PNCs and eBC mass concentration by inhibiting aerosol wet deposition.

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~~ss,aresthe . Firstly, the change of all three meteorological parameters agree perfect between urban and rural background sites. In the mountain area, more significant changes were observed in summer, autumn, and winterand ight by the changes of weather conditions. Second, the significant slope of precipitation was found only in summer, around -5.9 % per year. Decreased precipitation in summer might result in less wet deposition and thus in a smaller reduction rate of the eBC mass concentration and N[200-800]. For the ambient temperature, the significant increases were detected in summer, autumn and winter. Increased temperature, especially in winter, may lead to lower~~

~~anthropogenic emissions from domestic heating or power generation. In addition, slight increase of wind speed was observed in winter, resulting in an increased dilution and thus decreased pollutant concentrations.~~The relationship between the change of wind speed and pollutant parameters can be hardly seen from table 6. ~~In summary, increased ambient temperature and wind speed in winter might contribute to the decrease of the PNCs and eBC mass concentrations. However~~Conversely, ~~decreased precipitation in summer may diminish the decrease of the PNCs and eBC mass concentration.~~

4.2. Influence of air ~~-~~mass condition on the detected trends

Synoptic-scale air mass condition, including origin region and pathways is an important factor driving regional pollutant concentration (Ma et al., 2014; Hussein et al., 2006). ~~Besides, a~~Atmospheric stability is also important ~~since~~as it dominates the vertical dilution of pollutants. Based on a self-developed~~-~~ back-trajectory cluster method (BCLM), the influences of the inter-~~annal~~annual changes ~~of~~in air ~~-~~mass conditions and atmospheric stability on the detected trends are investigated.

~~**5~~4~~.2 Air mass dependency on long-term changes in particle number concentration and equivalent black carbon**~~

~~Synoptic-scale air masses, representing different weather conditions and long-range transport pattern, can be used to explain the different temporal variation of aged aerosol particles (Ma et al, 2014; Hussein et al., 2006). Two factors may control the concentration of aerosol particles in different air masses: residence time over the continent and regional emission at origin region.~~

~~To investigate the influence of the long-range transport pattern, a backward trajectory clustering method was used.~~ BCLM~~This method, denoted as back-trajectory cluster method (BCLM),~~ is based on a joint cluster analysis considering air ~~-~~mass backward trajectories~~, PM_{10} mass concentration~~, ~~and~~ profiles of pseudo-potential temperature, ~~and~~~~re~~ PM_{10} mass concentration ~~at several sites~~ over Germany~~, including regional background, low mountain range and high Alpine conditions~~ (Birmili et al., 2010; Engler et al., 2007; Ma et al., 2014). In ~~this study~~BCLM, 15 air mass types are ~~clustered~~defined ~~obtained from BCLM~~ to represent different ~~the overall~~ meteorological conditions~~,~~ on a ~~large~~ scale ~~over in~~of Germany~~, and it is thus valid for all GUAN sites~~. ~~It should be noticed that, the time span of BCLM is from 2009 to 2014, which does not totally cover the whole observation time in our trend analysis (2009-2018). However, as the trend evolution plots in Fig. 3 and one previous short-term study (2009-2013) of GUAN dataset (Birmili et al., 2015) shown, reductions of the PNCs and eBC mass concentration have been observed at most of GUAN sites during 2009-2014. Therefore, to evaluate the influence of long-range transport on the decrease of the PNCs and eBC mass concentration, the BCLM was used in this section since we believe the change of long-range transport pattern in 2009-2014 could represent its change in the whole time period (2009-2018).~~ ~~More~~Detailed information about data preparation, cluster processing, and the data procedures and data products is described ~~in detail~~ in ~~the supplementary~~Supplement ~~-~~Sect. 3 of SM and in ~~in a corresponding research article by~~ Ma et al. (2014). Figure ~~7~~ 8 ~~shows gives~~shows the average trajectories and the ~~average normalizsed~~normalized profiles of pseudo potential temperature (θ_v) for the 15 ~~each~~ air ~~-~~mass types. ~~According to their vertical stability and meteorological condition as shown in Fig. 9 (7b), t~~The 15 air mass types are named by ~~the~~ seasons (CS: cold season; TS: transition season; and WS: warm season) and synoptic patterns ~~atmospheric flow: CS: cold season; TS: transition season; WS: warm season; (~~(ST: Stagnant; A1: Anti-cyclonic ~~(A1 refers to the~~with air ~~-~~mass originating from Eastern Europe; ~~while~~ A2: Anti-cyclonic with air mass originating from ~~refers to the air mass from~~ west~~)~~; C1: cyclonic with air mass originating~~(Air masses~~ from relatively south~~-~~; ~~are named as C1~~C2: cyclonic with air mass originating from the north~~, whereas from north are named as C2~~)).

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~~The vertical stability is more stable at CS air masses and more neutral in WS air masses. In the same season and atmospheric flow,~~ Table ~~5~~ 6 lists the basic statistical information of ~~each of~~ the 15 air mass types.

Figure 9~~4~~ illustrates the statistics of PNCs and eBC mass concentrations for each air mass types at the regional background site category (MEL, WAL and NEU). Large differences in~~of~~ the mean values of $N_{[200-800]}$ and eBC mass concentrations are observed among different air -mass types; whereas~~ile~~ $N_{[10-30]}$ and $N_{[30-200]}$ show less significant difference as~~ince~~ $N_{[10-30]}$ and $N_{[30-200]}$ represent more local information and are not as sensitive as $N_{[200-800]}$ and eBC mass concentration~~to~~. In the following discussion, only $N_{[200-800]}$ and eBC mass concentration are used~~as the~~.

Due to the high sensitivity of $N_{[200-800]}$ and eBC mass concentration on the air mass types, frequency changes of air mass types might~~ay~~ lead to changes in the their long-term trends. However, it is difficult to investigate the influence for each air mass type since the frequencies of the air mass types are quite low (3.0 % – 12.0 %). ~~Accordingly~~Therefore, the 15 air -mass types are grouped into two categories according to pollution level. If both the median eBC mass concentration and $N_{[200-800]}$ are higher than their overall median concentration, the air -mass is grouped into polluted air -mass category~~.~~, and vice versa. ~~Accordingly, the 15 air mass types are grouped into two categories.According to eBC mass concentration levels, the 15 air mass types are grouped into two categories:~~

(1): Polluted air mass category that includes CS-ST, CS-A1, CS-A2, CS-C1, TS-A1, WS-ST, WS-A1, and WS-C1;

(2): Cleaner air mass category that includes CS-C2a, CS-C2b, TS-A2, TS-C1, TS-C2, WS-A2, and WS-C2.

The annual occurrence of polluted air mass category together with the annul mean values of $N_{[200-800]}$ and eBC mass concentration at the regional background and low mountain range sites are shown in Fig. ~~11~~910. No clear trend of the occurrence of polluted air mass category could~~an~~ be found. However,~~But~~ large difference is visible in the occurrences between different years. If the change in a~~a~~ir mass frequency ~~change~~ plays an important role in the variations in~~of~~ $N_{[20-0-800]}$ and eBC mass concentration, low polluted air mass occurrences should be associated with relatively low $N_{[200-800]}$ and eBC mass concentration. However, such a relationship is not visible in Fig. 910. ~~11~~ ~~()~~The annual mean values of $N_{[200-800]}$ and eBC mass concentration (black lines in Fig. 10) ~~consistently~~ decrease consistently. Therefore, it ~~can~~could be concluded that the inter-annual changes in~~of~~ synoptic-scale air -mass conditions ~~is~~ are not the reason for~~of~~ the decrease of ~~in~~pollutant concentrations shown in Sect. 3.~~Two factors may control the concentration of aerosol particles in different air masses: residence time over the continent and regional emission at origin region.~~

~~5~~4.2.1 ~~**Particle number concentration and equivalent black carbon mass concentration for each air mass type**~~

~~Figure 8 illustrates the median value of the PNCs and eBC mass concentrations with respect to the 15 air mass types at regional background site category (MEL, WAL and NEU). First, there is less significant difference on $N_{[10-30]}$ and $N_{[30-200]}$ among different air mass types, since $N_{[10-30]}$ and $N_{[30-200]}$ represent more local information. First, there is less significant difference on $N_{[10-30]}$ and $N_{[30-200]}$ among different air mass types, since $N_{[10-30]}$ and $N_{[30-200]}$ represent more local information. For the $N_{[200-800]}$ and eBC mass concentration, higher values are observed in CS air masses as shown in Fig. 8a and 8c. This can be explained by higher anthropogenic emissions and less~~

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~~dilution caused by lower PBL height in cold season. In the same season (WS, CS or TS), the median values of the $N_{[200-800]}$ and eBC mass concentration differ with regard to atmospheric air flows. The $N_{[200-800]}$ and eBC mass concentration at the air mass types A1 and ST (CS-A1, WS-A1, TS-A1, CS-ST, and WS-ST), are always higher than the ones at other air mass types in the same season. Because these air masses remained as least three days over Central Europe before reaching the measurement sites. During these three days, emitted aerosol particles are continuously accumulated into the air masses. Within these five air mass types, the median values of the $N_{[200-800]}$ and eBC mass concentration at CS-A1 and WS-A1 are relatively higher, since the anti-cyclonic air mass usually comes from Eastern Europe with more anthropogenic emissions. Moreover, the median values of the $N_{[200-800]}$ and the eBC mass concentration at the air masses type A2 (CS-A2, TS-A2, and WS-A2), C1 and C2 decrease steadily, which can be explained by the shorter residence time over the European continent.~~

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~~5.2.2 Influence of air masses frequency change on the trend of the particle number concentration and the equivalent black carbon mass concentration~~

~~As shown in Fig. 8, the PNCs and eBC mass concentration vary widely with respect to different air mass types, meaning the air mass type is one of the factors to change the pollutant concentrations, especially for aged accumulation mode particles $N_{[200-800]}$ and eBC mass concentration. Therefore, the frequency change of air masses might may lead to a change of long-term trend of the PNCs and eBC mass concentration. In this section, the relationship between air mass frequency change and concentration change is discussed.~~

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~~It is, however, hard to detect the long-term trend of pollutant parameters for each individual air mass type because the frequency of air masses varies in a range of 2.6 % to 12.4 % (see Table 5). This means that some of them are too sensitive to detect the temporal change since their frequencies are too low. Therefore, it is needed to further group the 15 air mass types. According to the different eBC mass concentration values at different air mass types (see Fig. 8a), the 15 different air mass types are grouped into two categories:~~

~~(1): Polluted air mass category includes CS-ST, CS-A1, CS-A2, CS-C1, TS-A1, WS-ST, WS-A1, and WS-C1;~~

~~(2): Cleaner air mass category includes CS-C2a, CS-C2b, TS-A2, TS-C1, TS-C2, WS-A2, and WS-C2.~~

~~Figure 9 shows the relationship between air mass frequency change and mean pollutant concentration change at all regional background and low mountain range sites, with respect to each air mass category. If the air mass frequency change is an dominate factor for the downward trend of BC and accumulation mode particle $N_{[200-800]}$, a decrease in polluted air mass frequency should be associated with a decrease in $N_{[200-800]}$ and eBC mass concentration. From Fig. 9a, the frequency of polluted air mass does not consistently decrease: It slightly decreased from 2009 to 2012, and then started to increase after 2012. However, the annual mean values of the PNCs and the eBC mass concentrations consistently decrease at both air mass categories for all parameters. Therefore, it can be concluded that the change of long-range transport pattern is not the reason causing the reduction of pollutant concentrations.~~

~~To sum up, the long-term change of meteorological parameters and long-range transport pattern are analyzed in this section to investigate their contribution to the downward trend of the PNCs and eBC mass concentration in Germany. The results show that increased ambient temperature and wind speed in winter since 2009 are thought to have a contribution to declined eBC mass concentration and PNCs, as a result of less anthropogenic emissions from domestic heating etc. and slightly stronger dilution by higher wind speed. However, decreased precipitation in summer may diminish the decrease of the PNCs and eBC mass concentration. Moreover, the change of air mass frequency was detected and the results indicate that the change of long-range transport pattern is not the factor~~

~~causing the reduction of pollutant concentrations. It is an indication that, the stringent emission mitigation policies in Germany and Europe have a beneficial effect on the declined eBC mass concentrations and PNCs.(Chen et al., 2016)~~

~~6~~5 Conclusions

In this ~~work~~~~stu~~study~~dy~~, long-term trends of atmospheric ~~particle number concentrations (~~PNCs~~)~~ and ~~the equivalent black carbon (~~eBC~~)~~ mass concentration over a 10~~-~~ years period (2009–~~-~~2018) ~~were~~ are determined for 16 sites ~~in the German Ultrafine Aerosol Network (GUAN), ranging from roadside to high Alpine environment distributed over Germany~~in the ~~German Ultrafine Aerosol Network (~~GUAN~~)~~, ranging from roadside to high Alpine environments. Overall, statistically significant ~~downward~~ decreasing trends ~~were~~ are found for ~~most of these~~85 % of the parameters and ~~observation sites~~~~environments~~observation sites. ~~-~~Concretely, ~~Concretely, t~~The annual ~~relative~~ slope of the ~~the~~ eBC mass concentration varies between −13.1 % and −1.7 % ~~per year~~~~yr-1~~per year. ~~And t~~The slopes of the PNCs vary from −17.2 % to −1.7 %, −7.8 % to −1.1 %, and −11.1 % to −1.2 % ~~per year yr-1~~per year ~~(only statistically significant trends)~~ for 10−30 nm, 30−200 nm, and 200−800 nm ~~particle diameter~~size ranges, respectively~~, the annual slopes of the eBC mass concentration of all 16 sites varies between -7.7 % and -1.8 % per year, and the significant slopes of the PNCs vary from -6.3 % to 2.7 %, -7.0 % to -2.0 %, and -9.5 % to -1.5 % per year for particles with diameters of 10-30 nm, 30-200 nm, and 200-800 nm, respectively. The regional Mann-Kendall test yielded regional-scale trends of eBC mass concentration, $N_{[30\text{-}200]}$ and $N_{[200\text{-}800]}$ of -3.8 %, -2.0 % and -2.4 %, respectively, indicating an overall decreasing trends for sub-micrometer PNC (except $N_{[10\text{-}30]}$) and eBC mass concentration all over Germany~~.

The regional Mann-Kendall test yields regional-scale trends of eBC mass concentration, $N_{[30–200]}$, and $N_{[200–800]}$ of –5 %, –2.5 % and –2.9 % per year, respectively, indicating an overall decreasing trend in sub-micrometre PNC (except $N_{[10–30]}$) and eBC mass concentration all over Germany.

Comparing the trends of measured parameters with the long-term change in total emission, we believe that the observed trends of PNCs and eBC mass concentrations are mainly due to the emission reduction. The decreasing trend of eBC mass concentration agrees well with the variation of BC total emission in Germany

~~The decreasing trends are found on the total emission of BC, PM2.5 and precursors, suggesting the From 2009 to 2017, BC total emission in Germany deceases about −3.4 % per year and highly agrees with the trend of mean eBC mass concentration~~, suggesting reduction in emissions is the dominant factor for the reduction in eBC mass concentrations over ~~Germany~~the country. The ~~measured~~decreasing rates of $N_{[30–200]}$, $N_{[200–800]}$, and $V_{[200–800]}$ ~~decreases about −2.5 % per year, which is~~are stronger than the decrease in the total emissions of all precursors and $PM_{2.5}$, which ~~. This discrepancy~~ could have been caused by the highly complex and nonlinear processes of secondary ~~particle~~aerosol formation. ~~Comparing the trends of measured parameters with total emission, we believe that the observed trends of eBC mass concentrations and PNCs are mainly due to the emission reduction.~~

The highest decrease in eBC mass concentration was observed during daytime ~~(06:00–18:00 local time)~~ at the roadside and urban background, implying a strong evidence of reduced traffic emissions in the urban area. When there are fewer motor vehicles at night, the PNCs and eBC mass concentration ~~and PNC~~ in ~~the city~~the urban sites also show a significant ~~decline~~decrease, which ~~also~~ could be due to the background concentration decrease caused by the reduction in other emissions such as domestic heating, industry, etc. Stronger reductions in PNCs are found

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in winter, which is ~~very~~ likely to be caused by ~~some other factors with strong seasonal variation, for example, the~~the decreased, emissions from domestic heating combustion~~.~~.

Meteorological conditions ~~and synoptic air mass conditions~~ are also able to influence the temporal variation of aerosol ~~particles~~concentration. The ~~influences of~~ inter-annual changes in precipitation~~,~~ and temperature~~, and wind speed~~ might have some influence on the detected trends by, indirectly, influencing anthropogenic emissions and inhibiting aerosol wet deposition, but they seem to be ~~limited and are~~ not the dominant factors for the ~~detected trends~~long-term decrease of the measured parameters. The influence of long-range transport pattern ~~on the long-term trend of measured parameters~~ is also evaluated and the inter-annual changes in synoptic-scale air mass conditions are found to be not the reason for the ~~reduction~~decrease in pollutant concentrations.

~~The ctions of varioussare found to bedecreasing trends ofThe diurnal and seasonal variation of the trends clearly show the effects of the mitigation policies of road transport and residential emissions. The influences of other factors such as airmasses, precipitation, temperature etc. were also examined and found to be less important or negligible. This study proves that a combination of emission mitigation policies can effectively improve the air quality on large spatial scales. It also suggests a long-term aerosol measurement network with multi-type sites is an efficient and necessary tool for the verification emission mitigation policies.~~

~~Given the relative novelty of the long-term measurements (particle number size distributions, BC) in a network such as GUAN, the results proved to be robust and comprehensive. Our study shows that long-term measurements of aerosol parameters in different environments can be instrumental in detecting and understanding the long-term effects of emission mitigation policies.Particularly, the highest regional decrease appears for the eBC mass concentration for which combustion processes from motor traffic and power generation are the major source in Germany. This implies that decreasing anthropogenic emissions might be one of the factors causing the reduction of the PNCs and eBC mass concentrations.~~

~~The highest decrease of eBC mass concentration was observed during both working days (from Monday to Friday) and daytime (06:00-18:00 LT) at roadside and urban background, which implies a strong evidence of reduced traffic emissions in urban area. As traffic volumes near those sites have changed little in comparison, our results are indicative of reductions in specific emission factors, facilitated e.g. by the introduction of diesel particle filters. At regional and mountain sites, most of the trends showed a constant decrease during the whole week and entire day, rather indicating that the sources for the decrease are far away from the regional background or mountains and closer to urban areas.~~

~~Meteorological conditions are also able to influence the temporal variation of aerosol particles. Seasonal trends show that the reduction of the PNCs and eBC mass concentrations occurs all year round, however, stronger in wintertime. There are three explanations for this result:~~

~~a) The influence of reduced anthropogenic emission on PNC is thought to be much more prominent in winter than in summer (Asmi et al., 2013),~~

~~b) Increased ambient temperature and wind speed in winter are also thought to have a contribution on declined eBC mass concentration and PNCs, as a result of less anthropogenic emissions from domestic heating etc. and stronger dilution,~~

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~~c) Decreased precipitation in summer might result in less wet deposition and thus less scavenging and a smaller reduction rate of eBC mass concentration and~~ ~~$N_{[200-800]}$.~~

~~Moreover, the change of air mass frequency was determined but the results indicate that the change of long-range transport pattern is not associated with the reduction of pollutant concentrations. The success of emission mitigation policies and decreased pollutant concentrations are highly associated with the reduced risk of human health and visibility. A decrease of 10 μg m~~ ~~$^{-3}$~~ ~~in the~~ ~~$PM_{2.5}$~~ ~~mass concentration was related with an increase of life expectancy of 0.61 ± 0.20 year for populations in 211 counties (Pope et al., 2009). The improved health effects because of decreased UPF and BC would be much greater compared with that of~~ ~~$PM_{2.5}$~~ ~~mass concentration. The reduction of these pollutant parameters could also significantly improve the visibility, especially in the urban area (Singh et al., 2017; Wan et al., 2011).As of 2018, 97 % of cities in low- and mid-income countries with more than 100,000 inhabitants do not meet the World Health Organization (WHO) air quality guidelines (WHO, 2018). The emission mitigation policies demonstrated in this article are significant for the decision makers to reduce the emissions and ambient human exposure to sub-micrometre particles.We therefore conclude that the declining anthropogenic emissions are the most likely decisive factor for the decrease of the eBC mass concentration and PNCs all over Germany.~~

This study suggests that a combination of emission mitigation policies can effectively improve the air quality on large spatial scales such as in Germany. Given the relative novelty of the long-term measurements (~~particle number size distributions~~PNSD, BC) in a network such as GUAN, the results proved to be robust and comprehensive. Our study ~~shows~~ also shows that long-term measurements of aerosol parameters in different environments could~~an~~ be very instrumental in detecting and understanding the long-term effects of emission mitigation policies.~~(Pope et al., 2009)(Singh et al., 2017;Wan et al., 2011)~~

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Table 1: Basic information ~~of~~ ~~about~~on the 16 ~~atmospheric measurement~~ sites in the German Ultrafine Aerosol Network (GUAN), in alphabetic order ~~(Birmili et al., 2016)*~~.

No.	Site name	Abbreviation	Status (Until 2017)	Site category	Elevation (m)	Location
1	Annaberg-Buchholz	ANA	In operation	Urban background	545 ~~m~~	50°34'18" N, 12°59'56" E
2	Augsburg	AUG	In operation	Urban background	485 ~~m~~	48°21'29" N, 10°54'25" E
3	Bösel	BOS	Terminated end of 2014	Urban background	17 ~~m~~	52°59'53" N, 07°56'34" E
4	Dresden-Nord	DDN	In operation	Roadside	116 ~~m~~	51°03'54" N, 13°44'29" E
5	Dresden-Winckelmann-straße	DDW	In operation	Urban background	120 ~~m~~	51°02'10" N, 13°43'50" E
6	Hohenpeißenberg	HPB	In operation	Low mountain range	980 ~~m~~	47°48'06" N, 11°00'34" E
7	Langen	LAN	In operation	Urban background	130 ~~m~~	50°00'18" N, 08°39'05" E
8	Leipzig-Eisenbahnstraße	LEI	In operation	Roadside	120 ~~m~~	51°20'45" N, 12°24'23" E
9	Leipzig-Mitte	LMI	In operation	Roadside	111 ~~m~~	51°20'39" N, 12°22'38" E
10	Leipzig-TROPOS	LTR	In operation	Urban background	126 ~~m~~	51°21'10" N, 12°26'03" E
11~~11~~	Melpitz~~Leipzig-West~~	MEL~~LWE~~	In operation~~Terminated end of 2016~~	Regional background~~Urban background~~	86~~122 m~~	51°31'32" N, 12°55'40" E~~51°19'05" N, 12°17'51" E~~
12~~12~~	Mülheim-Styrum~~Melpitz~~	MST~~MEL~~	In operation~~In operation~~	Urban background~~Regional background~~	37~~86 m~~	51°27'17" N, 06°51'56" E~~51°31'32" N, 12°55'40" E~~
13~~13~~	Neuglobsow~~Mülheim-Styrum~~	NEU~~MST~~	In operation~~In operation~~	Regional background~~Urban background~~	70~~37 m~~	53°08'28" N, 13°01'52" E~~51°27'17" N, 06°51'56" E~~
14~~14~~	Schauinsland~~Neuglobsow~~	SCH~~NEU~~	In operation~~In operation~~	Low mountain range~~Regional background~~	1205~~70 m~~	47°54'49" N, 07°54'29" E~~53°08'28" N, 13°01'52" E~~
15~~15~~	Waldhof~~Schauinsland~~	WAL~~SCH~~	In operation~~In operation~~	Regional background~~Low mountain range~~	75~~1205 m~~	52°48'04" N, 10°45'23" E~~47°54'49" N, 07°54'29" E~~
~~16~~	~~Waldhof~~	~~WAL~~	~~In operation~~	~~Regional background~~	~~75 m~~	~~52°48'04" N, 10°45'23" E~~
16~~17~~	Zugspitze (Schneefernerhaus)~~Zugspitze (Schneefernerhaus)~~	ZSF~~ZSF~~	In operation~~In operation~~	High alpine~~High alpine~~	2670~~2670 m~~	47°25'00" N, 10°58'47" E~~47°25'00" N, 10°58'47" E~~

~~*The site Leipzig-West (LWE) was not evaluated in this study and therefore, not shown in this table.~~

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Table 2: Technical details of the instrumentations of the 16 GUAN sites ~~instrumentations. Mobility particle size spectrometers (MPSS) follow the TROPOS design unless stated otherwise (Birmili et al., 2016)~~~~*~~.

NO.	Name	Type	Inlet height above ground (m)	Particle mobility size spectrometer type	Size range (nm)	eBC instrument	eBC cut-off
1	ANA	portable cabin	4 ~~m~~	MPSS	10–800 ~~nm~~	MAAP	PM_1
2	AUG	portable cabin	4 ~~m~~	D-MPSS	5–800 ~~nm~~	Aethalometer (Type 8100)	$PM_{2.5}$
3	BOS	portable cabin	4 ~~m~~	MPSS	10–800 ~~nm~~	MAAP	PM_{10}
4	DDN	portable cabin	4 ~~m~~	D-MPSS	5–800 ~~nm~~	MAAP	PM_1
5	DDW	portable cabin	4 ~~m~~	MPSS	10–800 ~~nm~~	MAAP	PM_1
6	HPB	building	12 ~~m~~	MPSS	10–800 ~~nm~~	MAAP	PM_{10}
7	LAN	portable cabin	14 ~~m~~	MPSS (TSI 3936)~~**~~	10–600 ~~nm~~	–	PM_1
8	LEI	building	6 ~~m~~	TDMPSS	5–800 ~~nm~~	MAAP	PM_1
9	LMI	portable cabin	4 ~~m~~	TDMPSS	5–800 ~~nm~~	MAAP	PM_{10}
10	LTR	portable cabin	16 ~~m~~	TDMPSS	5–800 ~~nm~~	MAAP	PM_{10}
11~~11~~	MEL~~LWE~~	portable cabin~~portable cabin~~	4~~4 m~~	D-MPSS~~TDMPSS~~	5–800~~10–800 nm~~	MAAP~~MAAP~~	PM_{10}~~PM_{10}~~
12~~12~~	MST~~MEL~~	portable cabin~~portable cabin~~	4~~4 m~~	MPSS (TSI 3936)~~D-MPSS~~	14–750~~5–800 nm~~	–~~MAAP~~	PM_{10}~~PM_{10}~~
13~~13~~	NEU~~MST~~	building~~portable cabin~~	6~~4 m~~	MPSS~~MPSS (TSI 3936)~~	10–800~~14–750 nm~~	MAAP~~–~~	PM_{10}~~PM_{10}~~
14~~14~~	SCH~~NEU~~	building~~building~~	6~~6 m~~	MPSS~~MPSS~~	10–800~~10–800 nm~~	MAAP~~MAAP~~	PM_{10}~~PM_{10}~~
15~~15~~	WAL~~SCH~~	building~~building~~	6~~6 m~~	MPSS~~MPSS~~	10–800~~10–800 nm~~	MAAP~~MAAP~~	PM_{10}~~PM_{10}~~
16~~16~~	ZSF ~~WAL~~	building~~building~~	6~~6 m~~	MPSS (TSI 3936)~~MPSS~~	~~1~~20–600~~10–800 nm~~	MAAP~~MAAP~~	PM_{10}~~PM_{10}~~
~~17~~	~~ZSF~~	~~building~~	~~6 m~~	~~MPSS (TSI 3936)~~	~~10–600 nm~~	~~MAAP~~	~~PM_{10}~~

~~*The site LWE was not evaluated in this study and therefore, not shown in this table.~~

~~** TSI 3031 has been measured in parallel at LAN for 2015–2018 (20–500 nm).~~

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Table ~~3~~33: Multi-annual trends of the PNCs and eBC mass concentration ~~and PNCs~~ in percentage per year, calculated using the ~~customized~~ customised Sen's estimator and generalized ~~linear~~ least squares regression with autoregression bootstrap (GLS /ARB). ~~The bold slopes are the s~~Statistically significant slopes at ~~the~~ 95 % significance level ~~in % per year~~are marked as bold numbers. Five site categories ~~on~~ in the left column are roadside (RS), urban background (UB), regional background (RB), low mountain range (LMT), and high Alpine (HA).

Category	Site	eBC mass concentration		$N_{[20\text{-}800]}$		$N_{[10\text{-}30]}$		$N_{[30\text{-}200]}$		$N_{[200\text{-}800]}$	
		Sen slope	GLS /ARB slope	Sen slope	GLS /ARB slope	Sen slope	GLS /ARB slope	Sen slope	GLS /ARB slope	Sen slope	GLS /ARB slope
RS	DDN	**−~~-~~11.3~~%~~**	**−13.1~~-13.1%~~**	**−7.3~~-7.3%~~**	**−6.0~~-4.4%~~**	**−~~-~~8.0~~%~~**	**−7.3~~-6.8%~~**	**−~~-~~6.7~~%~~**	**−5.6~~-4.4%~~**	**−~~-~~9.7~~%~~**	**−8.6~~-8.7%~~**
	LEI	**−~~-~~5.0~~%~~**	**−6.3~~-6.7%~~**	**−2.9~~-2.9%~~**	**−4.0~~-3.4%~~**	**−~~-~~5.0~~%~~**	**−4.8~~-4.6%~~**	**−~~-~~2.9~~%~~**	**−3.8~~-3.4%~~**	**−~~-~~1.2~~%~~**	**−3.3~~-2.5%~~**
	LMI	**−~~-~~5.5~~%~~**	**−6.8~~-6.4%~~**	**−4.8~~-4.8%~~**	**−6.9~~-7.3%~~**	−~~-~~0.2~~%~~	**−1.4~~0.0%~~**	**−~~-~~5.5~~%~~**	**−7.3~~-7.3%~~**	**−~~-~~4.8~~%~~**	**−6.7~~-6.7%~~**
UB	MST	---	---	**−~~-~~2.6~~%~~**	**−1.5~~-0.5%~~**	---	---	**−~~-~~3.2~~%~~**	**−1.8~~-0.5%~~**	**−~~-~~6.1~~%~~**	**−5.6~~-5.4%~~**
	LTR	**−~~-~~4.1~~%~~**	**−6.0~~-6.4%~~**	**−~~-~~4.3~~%~~**	**−6.7~~-5.5%~~**	**−~~-~~4.7~~%~~**	**−17.2~~-6.7%~~**	**−~~-~~4.1~~%~~**	**−5.4~~-5.5%~~**	**−~~-~~4.6~~%~~**	**−5.2~~-9.9%~~**
	ANA	**−~~-~~6.9~~%~~**	**−13.1~~-9.7%~~**	**−~~-~~5.5~~%~~**	**−7.7~~-6.8%~~**	**−~~-~~6.5~~%~~**	**−8.4~~-7.1%~~**	**−~~-~~5.4~~%~~**	**−7.5~~-6.8%~~**	**−~~-~~11.1~~%~~**	**−8.2~~-7.6%~~**
	AUG	**−~~-~~2.3~~%~~**	**−3.9~~-3.8%~~**	**−~~-~~6.3~~%~~**	**−7.9~~-9.7%~~**	**−~~-~~6.0~~%~~**	**−8.8~~-8.7%~~**	**−~~-~~6.3~~%~~**	**−7.8~~-9.7%~~**	**−~~-~~3.7~~%~~**	**−5.1~~-4.9%~~**
	DDW	**−~~-~~8.1~~%~~**	**−11.0~~-11.5%~~**	**−~~-~~4.8~~%~~**	**−8.9~~-9.2%~~**	**−~~-~~3.2~~%~~**	**−5.8~~-8.4%~~**	**−~~-~~5.0~~%~~**	**−5.4~~-9.2%~~**	**−~~-~~8.8~~%~~**	**−8.2~~-9.1%~~**
	LAN	---	---	**−~~-~~3.4~~%~~**	**−2.6~~-3.0%~~**	**−~~-~~1.4~~%~~**	−1.1~~-0.6%~~	**−~~-~~4.3~~%~~**	**−3.4~~-3.0%~~**	**−~~-~~2.5~~%~~**	−2.0~~-1.0%~~
	BOS	**−~~-~~4.9~~%~~**	**−6.2~~-6.6%~~**	**−~~-~~5.5~~%~~**	**−5.5~~-5.5%~~**	**−~~-~~1.7~~%~~**	**−3.9~~-5.8%~~**	**−~~-~~5.9~~%~~**	**−6.0~~-5.5%~~**	**−~~-~~6.3~~%~~**	−3.6~~-2.9%~~
RB	MEL	**−~~-~~4.4~~%~~**	**−7.6~~-7.4%~~**	−~~-~~0.2~~%~~	**−3.5~~0.1%~~**	1.9~~%~~	0.4~~1.6%~~	−~~-~~0.2~~%~~	−0.3~~0.1%~~	**−~~-~~2.9~~%~~**	**−3.8~~-6.4%~~**
	WAL	**−~~-~~3.2~~%~~**	**−3.7~~-4.7%~~**	**−~~-~~4.2~~%~~**	**−3.6~~-3.5%~~**	**−~~-~~3.3~~%~~**	**−3.5~~-2.4%~~**	**−~~-~~4.4~~%~~**	**−3.7~~-3.5%~~**	**−~~-~~5.2~~%~~**	**−6.2~~-5.7%~~**
	NEU	**−~~-~~7.8~~%~~**	**−7.8~~-8.3%~~**	−~~-~~1.0~~%~~	−0.2~~-0.6%~~	−~~-~~0.6~~%~~	−0.3~~0.2%~~	−~~-~~0.5~~%~~	0.0~~-0.6%~~	**−~~-~~3.9~~%~~**	**−4.8~~-5.5%~~**
LMT	HPB	**−~~-~~2.8~~%~~**	**−6.3~~-6.3%~~**	**−~~-~~1.2~~%~~**	**−3.1~~-2.6%~~**	**1.7~~%~~**	−0.6~~0.3%~~	**−~~-~~1.2~~%~~**	**−1.1~~-2.6%~~**	**−~~-~~3.9~~%~~**	**−5.6~~-6.0%~~**
	SCH	**−~~-~~1.7~~%~~**	**−3.6~~-4.2%~~**	**−~~-~~1.5~~%~~**	**−3.0~~-3.3%~~**	**3.8~~%~~**	−1.9~~-3.1%~~	**−~~-~~2.0~~%~~**	**−3.0~~-3.3%~~**	**−~~-~~3.8~~%~~**	**−3.8~~-4.5%~~**
HA	ZSF	**−~~-~~4.0~~%~~**	**−6.6~~-7.6%~~**	**−~~-~~4.2~~%~~**	**−4.9~~-5.5%~~**	---	---	**−~~-~~4.1~~%~~**	**−4.3~~-5.5%~~**	**−~~-~~4.2~~%~~**	**−9.7~~-11.3%~~**
Regional Mann-Kendall		**−~~-~~5.0~~%~~**		**−~~-~~2.1~~%~~**		−~~-~~1.4~~%~~		**−~~-~~2.5~~%~~**		**−~~-~~2.9~~%~~**	

Table 4. Comparison of long-term trend studies of BC, PNC, and PM in Europe.

Study	**Time period**	**Region**	**Parameters**	**Annual slope** (~~* slopes in the~~ numbers in brackets ~~is~~are the absolute slope, in µg m^{-3} yr^{-1})
This study	2009–2018	Germany	BC	Traffic (3 sites): −11.3 %~−5.0 %, (−0.19~~~,~~−0.08)~~, 3 sites~~; UB (5 sites): −8.1 %~−2.3 % (−0.08~~~,~~−0.03)~~, 5 sites~~; RB to high Alpine (6 sites): −7.8 %~−1.7 % (−0.03~~~,~~0.00)~~, 6 sites.~~
			$N_{[20\text{-}800]}$	Traffic (3 sites): −7.3 %~−2.9 %; UB (7 sites): −6.3 %~−2.6 %~~, 7 sites~~; RB to high Alpine (6 sites): −4.2 %~−0.2 %~~, 6 sites.~~
Kutzner et al., 2018	2005–2014	Germany	BC	Traffic (7 sites): (−0.31, −0.15)~~, 7 sites~~; UB (4 sites): (−0.1, −0.02)~~, 4 sites~~; Rural (1 site): 0.00~~, 1 site~~
Asmi et al., 2013	2001–2010	Europe	$N_{[20\text{-}800]}$	Rural to remote (4 sites): −4.6 %~1.6 %~~, 4 sites~~
Collaud Coen et al., 2013	2001–2010	Europe	Absorption coef.	Rural to remote (4 sites): −1.6 %~0.0 %~~, 4 sites~~
Bigi and Ghermandi, 2016	2005–2014	Italy, Po valley	$PM_{2.5}$	Traffic (2 sites): −6.4 %~−4.6 %~~, 2 sites~~; UB (17 sites): −8.1 %~ −0.4 %~~, 17 sites~~; RB (4 sites): −4.9 %~0.0 %~~, 4 sites~~
Singh et al., 2018	2009–2016	United Kingdom	BC	Traffic (1 site): −8.0 %~~, 1 site~~; UB (2 sites): −5.0 %~−4.7 %~~, 2 sites~~; Rural (1 site): −7.7 %~~, 1 site~~

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Table ~~4~~5: Trends of meteorological parameters for the three site categories ~~all over~~ in Germany. The bold numbers are the statistically significant slopes at the 95 % significance level. The daily meteorological data are from Germany's National Meteorological Service (Deutscher Wetterdienst, DWD).~~s The mean time series among for all 76 sitesthree site categories was used as the area average of meteorological parameters all over Germany.~~

~~s~~		**Urban background**	**Regional background**	**Mountain area**
Spring	Precipitation mm $year^{-1}$ (% $year^{-1}$)	−0.02 (−1.0)	0.00 (0.0)	−0.02 (−0.5)
	Temperature °C $year^{-1}$ ~~(% $year^{-1}$)~~	−0.04 ~~(−0.5)~~	−0.03 ~~(−0.3)~~	−0.02 ~~(−0.7)~~
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	0.01 (0.2)	0.02 (0.3)	0.04 (0.6)
Summer	Precipitation mm $year^{-1}$ (% $year^{-1}$)	**−0.14 (−5.5)**	**−0.15 (−5.8)**	**−0.20 (−4.7)**
	Temperature °C $year^{-1}$ ~~(% $year^{-1}$)~~	**0.15** ~~**(0.8)**~~	**0.13** ~~**(0.7)**~~	**0.16** ~~**(1.4)**~~
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	0.00 (0.0)	0.02 (0.4)	**−0.08 (−1.4)**
Autumn	Precipitation mm $year^{-1}$ (% $year^{-1}$)	−0.07 (−3.9)	−0.05 (−2.5)	**−0.07 (−1.9)**
	Temperature °C $year^{-1}$ ~~(% $year^{-1}$)~~	**0.37** ~~**(3.6)**~~	**0.36** ~~**(3.4)**~~	**0.29** ~~**(5.9)**~~
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	−0.02 (−0.8)	−0.01 (−0.3)	**−0.09 (−1.2)**
Winter	Precipitation mm $year^{-1}$ (% $year^{-1}$)	0.02 (1.3)	0.04 (1.8)	**0.14 (3.1)**
	Temperature °C $year^{-1}$ ~~(% $year^{-1}$)~~	**0.41** ~~**(29.9)**~~	**0.43** ~~**(23.2)**~~	**0.34** ~~**(Increase)**~~
	Wind speed m s^{-1} $year^{-1}$ (% $year^{-1}$)	0.02 (0.5)	0.05 (0.9)	**0.13 (1.5)**

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Table ~~5~~6: Basic statistical information of the 15 ~~different~~ air mass types.

Air mass type	Wind direction	Source region	Frequency 2009– ~~-2014~~ 2018 (%)	Mean PM_{10} 2009–2018 (μg m^{-3})
CS-ST	Stagnant	Central Europe	3.0~~2.6~~	34.6~~39.6~~
CS-A1	East	Eastern Europe	3.4~~4.0~~	34.8~~36.5~~
CS-A2	West	North Atlantic	5.8~~5.6~~	23.1~~25.6~~
CS-C1	South West	Southwest Europe	5.3~~5.2~~	24.4~~26.6~~
CS-C2a	South West	North Atlantic	4.0~~3.6~~	11.5~~12.8~~
CS-C2b	West	North Atlantic	5.8~~5.5~~	11.3~~13.0~~
TS-A1	North East	Subpolar	7.4~~8.3~~	17.5~~19.8~~
TS-A2	West	North Atlantic	6.5~~6.3~~	16.9~~18.7~~
TS-C1	South West	Southwest Europe	5.0~~5.1~~	14.4~~15.4~~
TS-C2	North West	Arctic	10.2~~10.8~~	12.9~~14.1~~
WS-ST	Stagnant	Central Europe	7.4~~6.8~~	20.5~~23.2~~
WS-A1	South East	Eastern Europe	5.9~~5.6~~	24.8~~28.4~~
WS-A2	North West	North Atlantic	12.0~~12.4~~	16.7~~17.9~~
WS-C1	West	North Atlantic	9.1~~9.7~~	16.4~~18.0~~
WS-C2	West	North Atlantic	9.0~~8.3~~	12.1~~13.0~~

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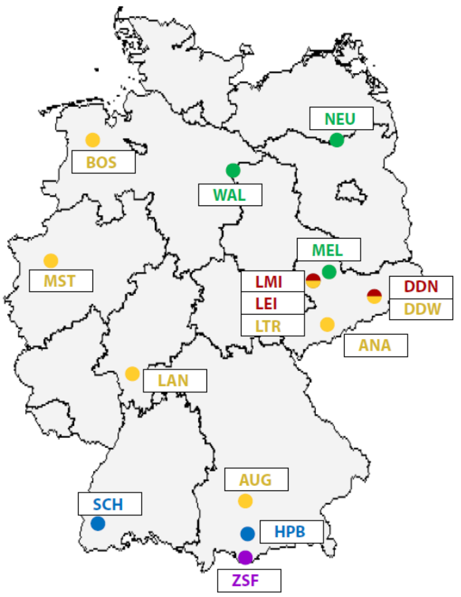
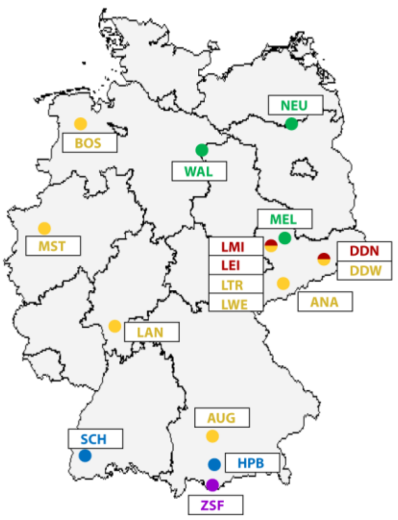


Abbreviation	Site name
ANA	Annaberg-Buchholz
AUG	Augsburg
BOS	Bösel
DDN	Dresden-Nord
DDW	Dresden-Winckelmannstraße
HPB	Hohenpeißenberg
LAN	Langen
LEI	Leipzig-Eisenbahnstraße
LMI	Leipzig-Mitte
LTR	Leipzig-TROPOS
MEL	Melpitz
MST	Mülheim-Styrum
NEU	Neuglobsow
SCH	Schauinsland
WAL	Waldhof
ZSF	Zugspitze (Schneefernerhaus)

City with roadside and urban background sites
Urban background sites
Regional background sites
Low mountain range sites
High Alpine site



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LMI	Leipzig-Mitte
LTR	Leipzig-TROPOS
LWE	Leipzig-West
MEL	Melpitz
MST	Mülheim-Styrum
NEU	Neuglobsow
SCH	Schauinsland
WAL	Waldhof
ZSF	Zugspitze (Schneefernerhaus)

City with roadside and urban background sites
Urban background sites
Regional background sites
Low mountain range sites
High Alpine site

Figure 1: ~~The m~~Map of the 16 atmospheric measurement stations in the GUAN.

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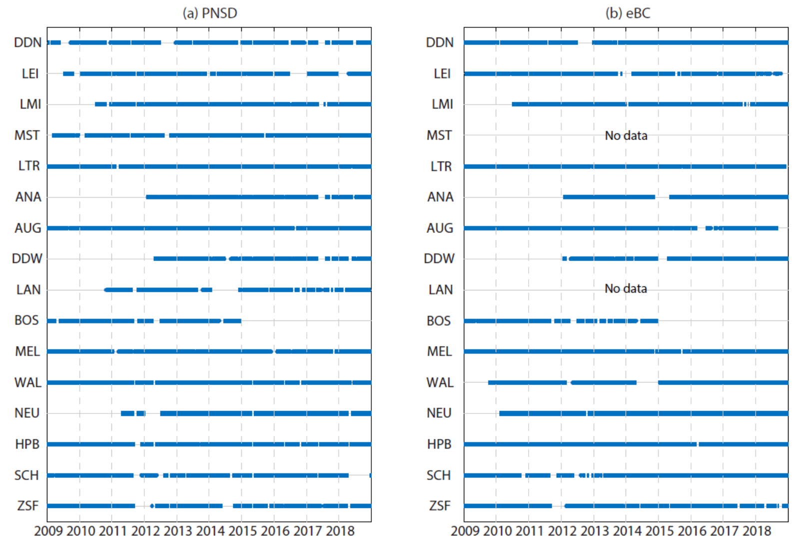

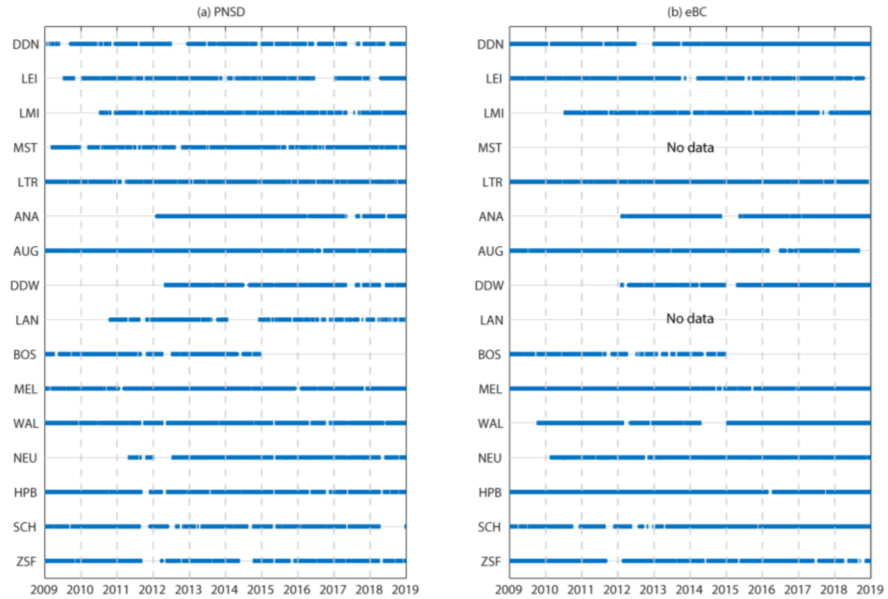

Figure 2: Data coverage of the PNSD and ~~the~~ eBC mass concentrations from 2009 to 2018 at the 16 GUAN sites~~, from 2009 to 2018~~.

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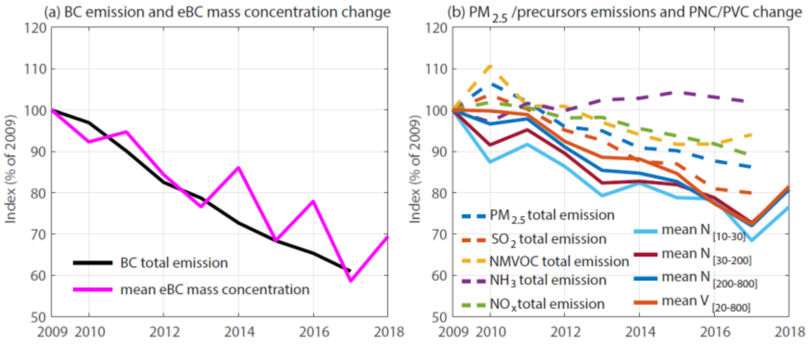

Figure 3: Comparison ~~between~~of the ~~changes~~long-term changes in measured parameters and total emissions in Germany.

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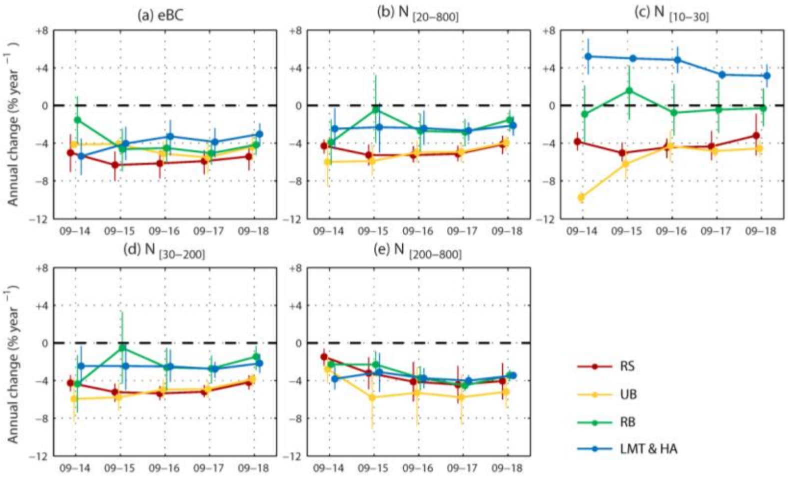

~~**Figure 3: Annual trends of the eBC mass concentration and PNCs for expanding time intervals starting from 2009, using the customized Sen's estimator. The x-axis shows the starting and ending year of each data point. The dot indicates the mean slope and the whiskers denote the 75th and 25th percentiles. The trend evolution for each site category is illustrated: roadside (RS), urban background (UB), regional background (RB), low mountain range and high Alpine (LMT&HA).**~~

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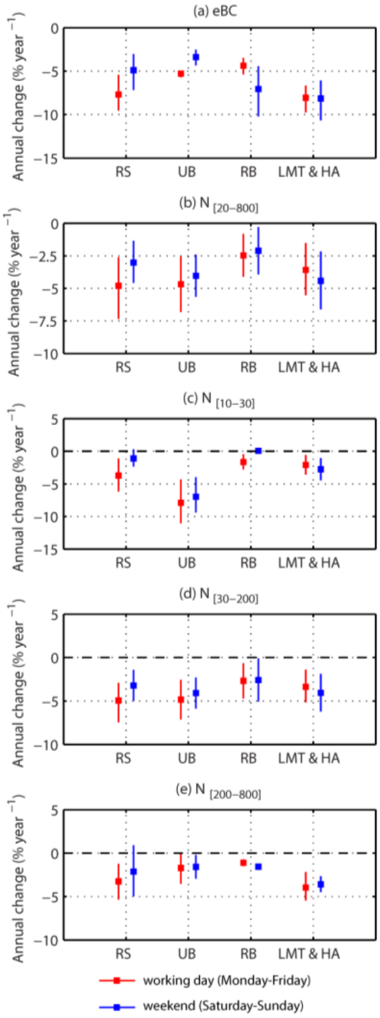

~~Figure 4: Annual trends of the eBC mass concentration and PNCs for working days and weekend, using the customized Sen's estimator at each site category: roadside (RS), urban background (UB), regional background (RB), low mountain range and high Alpine (LMT&HA). The square denotes the average Sen's slope on corresponding days (working day or weekend) and the whiskers denote the 25^{th} and 75^{th} percentile of Sen's slopes.~~

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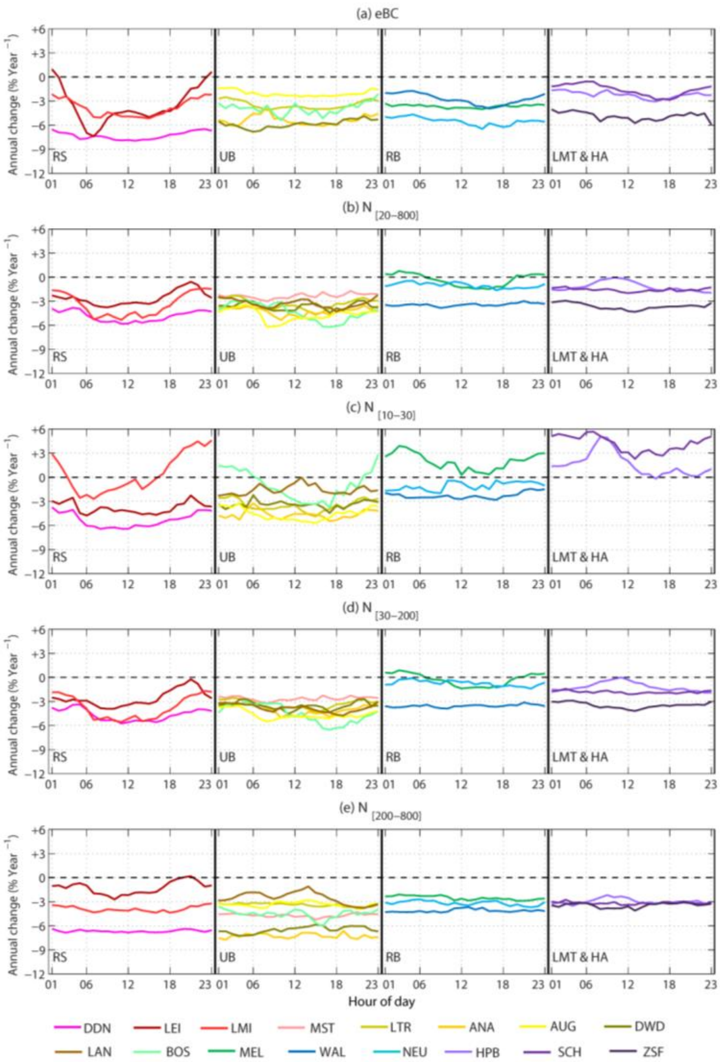
(a) eBC
(b) N [20–800]
(c) N [10–30]
(d) N [30–200]
(e) N [200–800]
Annual change (% Year −1)
+6
+3
0
−3
−6
−9
−12
RS
UB
RB
LMT & HA
01
06
12
18
23
Hour of day
DDN
LEI
LMI
MST
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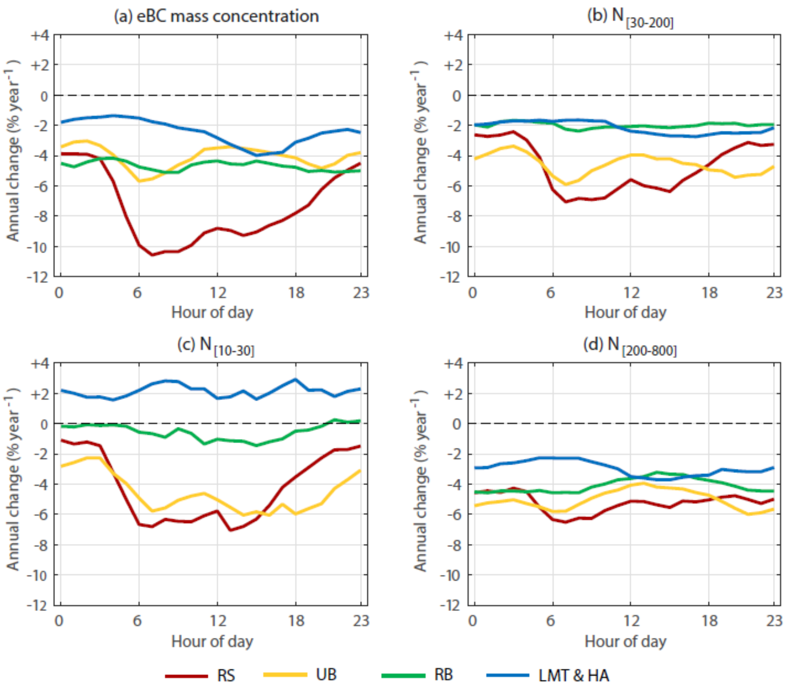

Figure ~~5~~4: Diurnal variations of the ~~Multi-annual~~ trends of ~~the~~ PNCs and eBC mass concentration ~~and PNCs corresponding to each hour of the day~~ for each site category~~,~~ ~~based on the customized customised Sen's estimator~~.

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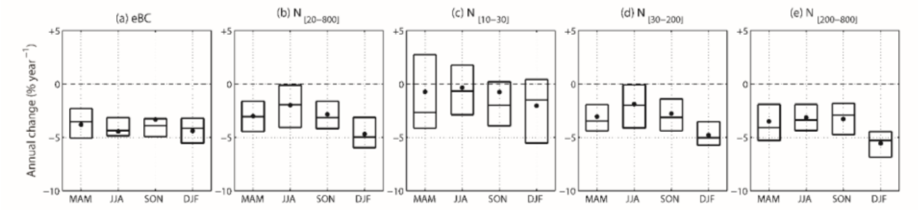

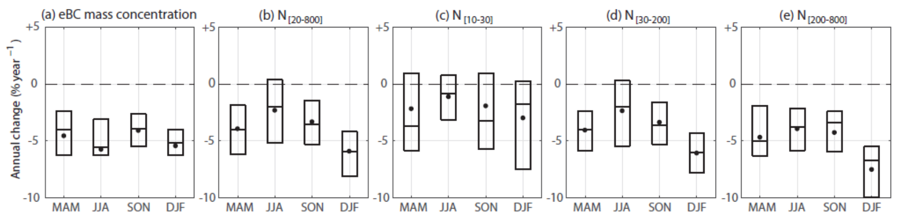

Figure ~~6~~5: Seasonal statistics of ~~annual~~ the trends of PNCs and ~~the~~ eBC mass concentrations ~~and PNC,s, based on the customized customised Sen's estimator:~~ Dots denote the mean slope for all sites, black line inside the box denotes the median slope, and the top and bottom of the box denotes the 75th and 25th percentiles. Spring: March to May (MAM); summer: June to August (JJA); autumn: September to November (SON); and winter: December to February (DJF). ~~Dots refer to mean slope for allat all sites, black line inside the box refers to the median slope, and the top and bottom of the box denotes the 75th and 25th percentiles.~~

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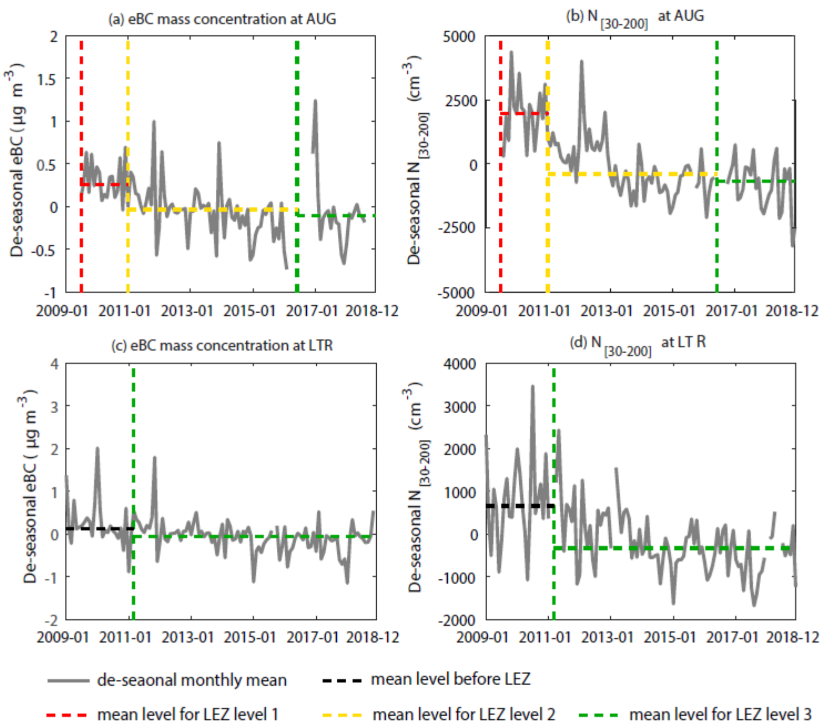

Figure 6: De-seasonalised monthly time series of eBC mass concentration and $N_{[30-200]}$ at the two urban background sites AUG and LTR. The vertical dashed lines refer to the start dates of LEZ ~~introduce dates~~of ~~, of~~ different stages in the city of Augsburg and Leipzig. The horizontal dashed lines refer to the mean concentration levels of measured parameters during the corresponding time period.

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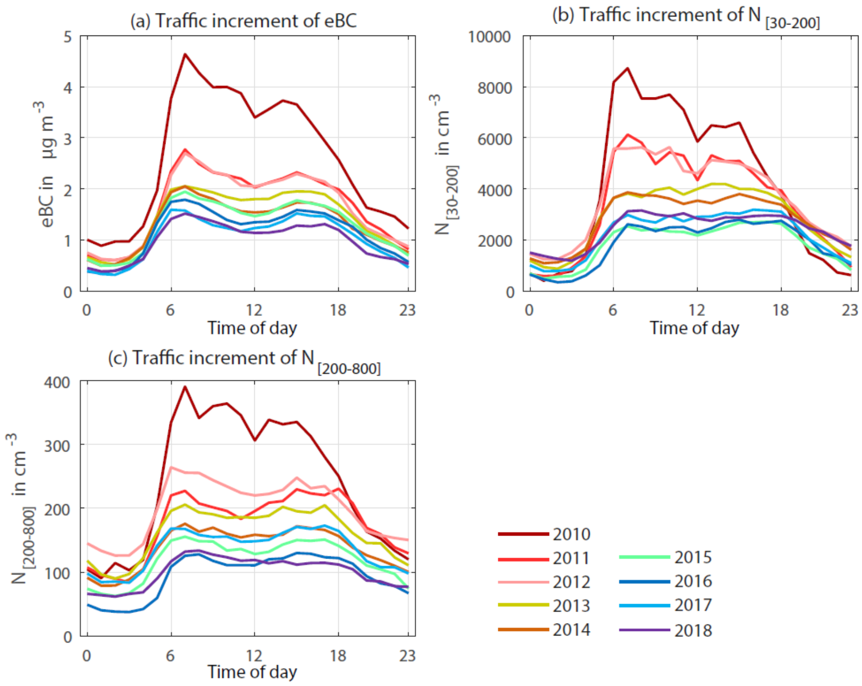

(a) Traffic increment of eBC
eBC in μg m-3
Time of day
(b) Traffic increment of N [30-200]
N[30-200] in cm-3
Time of day
(c) Traffic increment of N [200-800]
N[200-800] in cm-3
Time of day
2010
2011
2012
2013
2014
2015
2016
2017
2018

1572

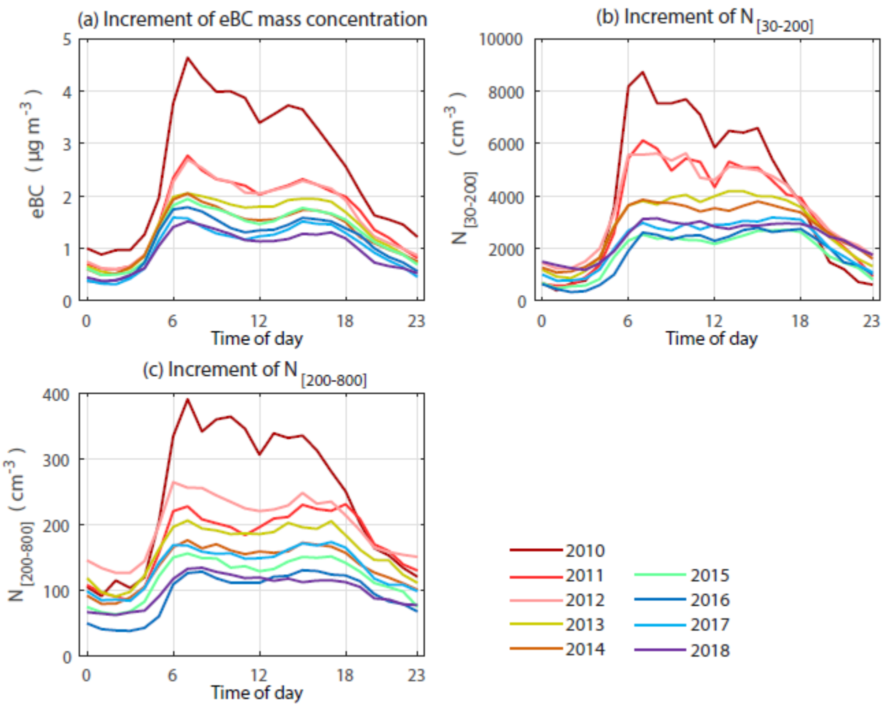

Figure 7: Average ~~7: Weekly~~ diurnal cycles of the increment (defined as the difference between LMI and LTR) in eBC mass concentrations, $N_{[30\text{-}200]}$ and $N_{[200\text{-}800]}$ ~~and PNCs for the traffic increment condition, defined as the different concentrations between LMI and LTR~~.

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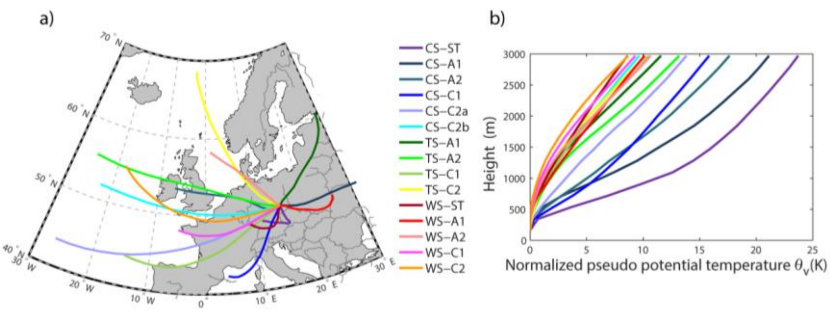

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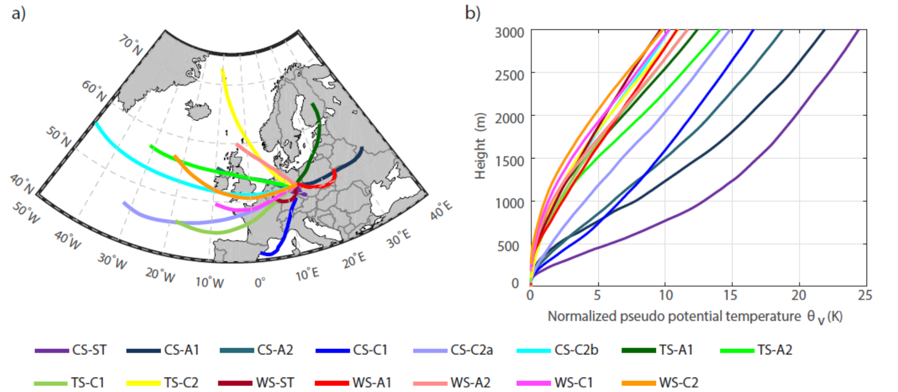

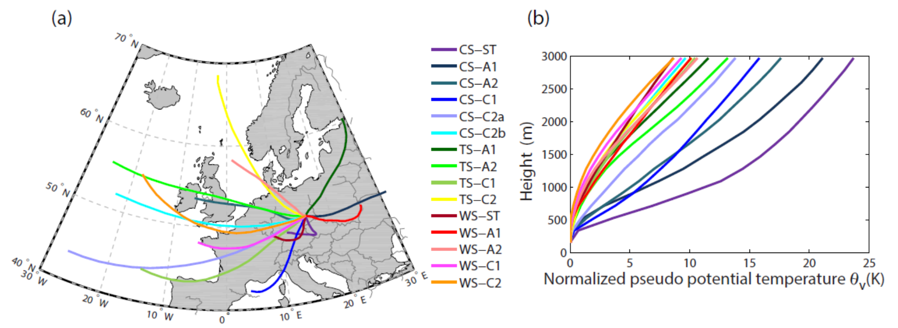

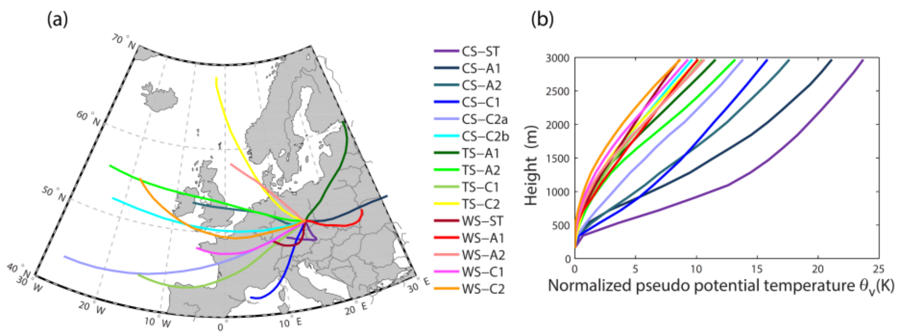

Figure ~~7~~8: ~~Basic information on the back-trajectory cluster model (BCLM).~~ (a)~~:~~ 15 back-trajectory clusters ~~centers~~ ~~centres~~ terminated at MEL ~~as an example~~. ~~(The duration of the back trajectories is 72h. The name of each air mass cluster refers to the character of each cluster: CS: cold season; TS: transition season; WS: warm season; ST: Stagnant; A: Anticyclonic; C: Cyclonic.~~ b) Average ~~normalized~~ normalised profiles of pseudo potential temperature (θ_v) for the 15 ~~air mass~~ clusters. ~~Profiles with a flat gradient indicate temperature inversions, while a steep gradient, imply stratification close to neutral. Data originate from the radiosounding launched at the DWD station Lindenberg, located 115 km northeast of MEL.~~

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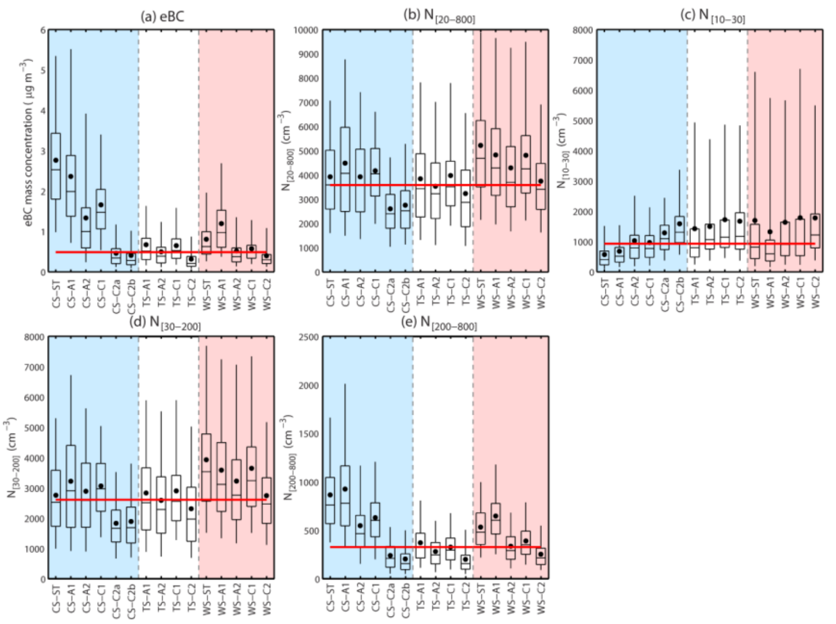

~~Figure 8: Average concentration value of eBC mass concentration (a) and size-dependent PNCs (b to e) for the 15 air mass types at regional background site category (Sites MEL, WAL and NEU). For each panel, the boxes and whiskers denote the 5th, 25th, 50th, 75th and 95th percentiles, while the dots denote the mean values. The solid red line indicates the overall median values.~~

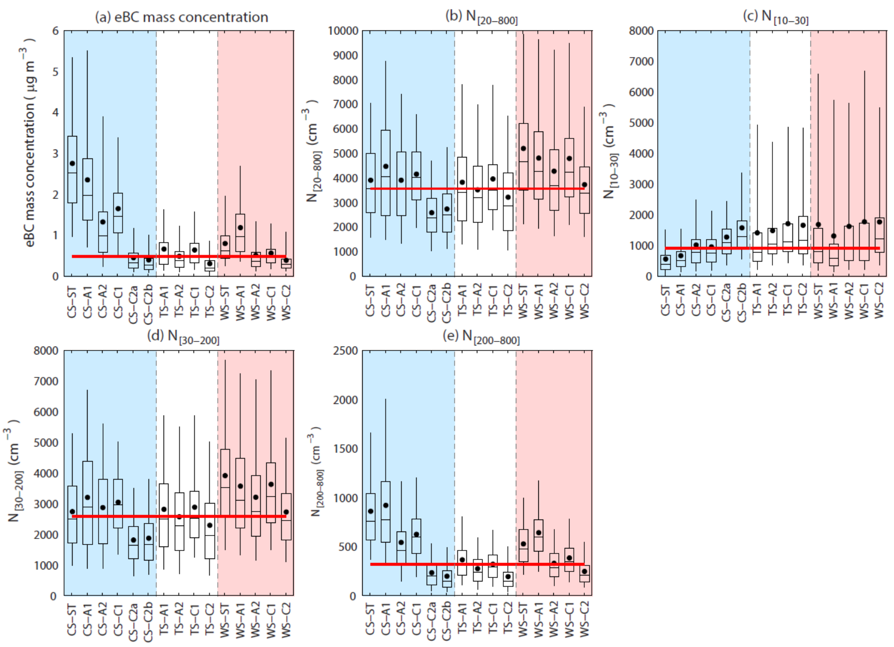

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Figure 9: Average concentrations of eBC mass concentration (a) and PNCs (b to e) for the 15 air mass types at the regional background site category (MEL, WAL and NEU). In each panel, the boxes and whiskers denote the 5th, 25th, 50th, 75th and 95th percentiles, while the dots denote the mean values. The solid red line shows the overall median values.

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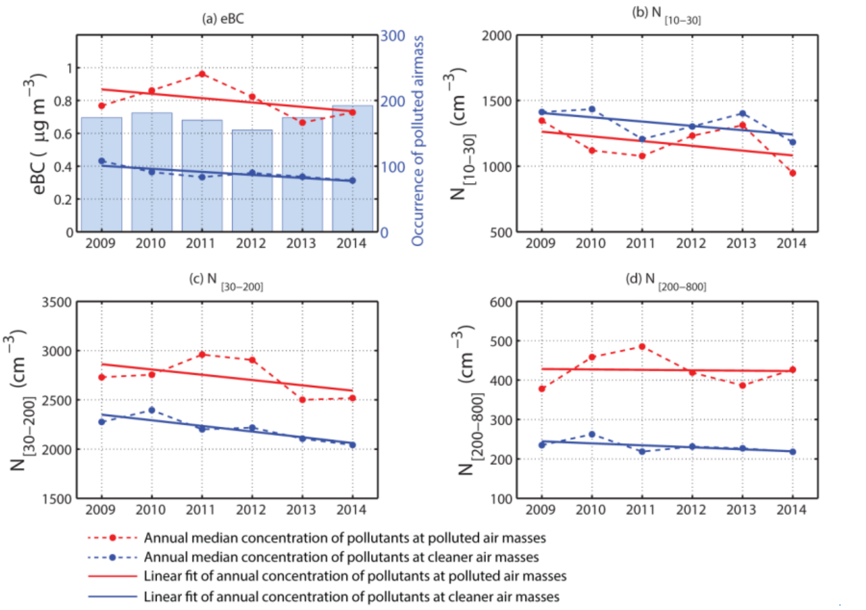

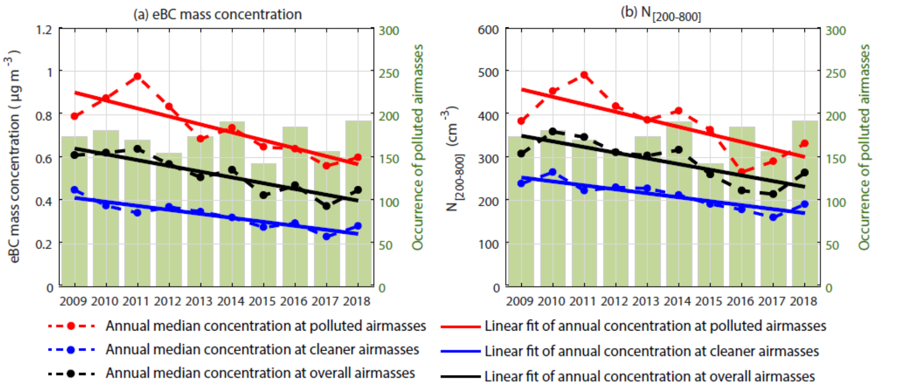

Figure 910 ~~9~~: Annual concentrations of the eBC mass concentrations and ~~*PNCs*~~ $N_{[200-800]}$ for the ~~two~~ polluted and clean air mass categories. Green bars show the ~~s, and~~ frequency of polluted air masses in each year.

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