30-Jan-2026

Dear Referees CR-1 and CR-2,

herewith we give our response to your valuable comments:

CR-1: Several sentences exceed 30–40 words. Breaking them into two improves readability.

AC: We have revised the text and found long sentences in the following lines, which we divided up and shortened in a meaningful way: 28, 66, 111, 116, 241, 269, 341,376,383,423,433

CR-1: The introduction is thorough but could be more concise. Consider tightening the literature review by focusing more directly on gaps your study addresses.

AC: Thank you for this suggestion. We have rewritten the introduction in order to improve coherence and readability. We narrowed the literature review towards the specific gaps addressed in our study. Paragraphs were merged if possible, and now better connect with each other. We refer the referees to the revised version of the manuscript:

“Atmospheric aerosol and air quality science have achieved major advances in recent years, but important questions remain. Specific challenges concern the sources of ultrafine particles (UFP; particles with a size < 100 nm in diameter), their further processing in the atmosphere and finally, their impacts on climate and human health. The adverse health effects of UFP have not been fully ascertained, mostly due to a lack of sufficient long-term studies (Cassee et al., 2019; Ohlwein et al., 2019). Nevertheless, the WHO has forwarded concrete atmospheric UFP concentrations as a “good practice statement” in its latest report (WHO, 2021). It is worth to note that WHO defines UFP as a total particle number concentration (TNC) with a lower size limit of 10 nm or less and an open upper end. The WHO considers a 24-hour mean of TNC < 1,000 cm^-3 as “low concentrations”. Values of TNC > 10,000 cm^-3 are classified as “high concentrations”. The new EU Air Quality Directive 2024/2881 (Anon, 2024) includes the requirement to measure Particle Number Concentration (PNC) and Particle Size Distribution (PSD).

Atmospheric UFP and the particle number size distributions (PNSD) may occur in considerable variation in the atmosphere due to rapid dynamic processes such as particle nucleation from gas-phase compounds, subsequent particle growth, coagulation, deposition, and activation as cloud condensation nuclei (CCN) (Bousiotis et al., 2021; Yao et al., 2018). As shown by stationary and mobile observations, large and frequent spatio-temporal gradients occur in urban atmospheres (Trechera et al., 2023; von Schneidemesser et al., 2019). The overall factors influencing UFP concentrations include time of day, wind direction, season, wind speed, temperature and solar radiation (von Bismarck-Osten et al., 2013). The mixed layer height (MLH) (Emeis et al., 2008) is a critical factor in the dispersal of any kind of pollutant near the ground. Over continental areas, the mixed layer is typically more shallow at night-time and during the winter season (Ma, 2013; Geiss, 2017), effectively trapping pollutants emitted near the ground.

Combustion is a major source of UFP including a range of individual sources such as the engines of vehicles, aircraft, and ships, as well as power and heat generation plants and domestic heating. Airborne measurements suggest that plumes from industrial plants contribute to 10-40% of background UFP in Germany (Junkermann et al., 2016). The effects of road traffic are evidenced by numerous studies in urban areas (e.g., Wehner et al., 2002; Gani et al., 2021).

A contrasting phenomenon is secondary new particle formation (NPF), i.e., nucleation from gaseous precursors and subsequent growth into larger diameters. This process is typically

driven by the photochemical generation of the combination of one or more of the following compounds: sulphuric acid, amines and organic molecules. This may occur simultaneously over large spatial areas and is independent from combustion-driven UFP emission (Kerminen et al., 2018). In a continental atmosphere, combustion-derived UFP and (secondary) NPF may overlap in observations, making a separation of the contributions difficult. Ma and Birmili (2015) attempted to separate these contributions based on multiple-site observations and concluded that secondary NPF contributed up to 30% of nucleation mode particles in an urban area. Clustering analysis identified NPF as the most important source for 16% of particle size distributions in Southern Europe (Brines et al., 2015). Several studies have intensely discussed the source apportionment of UFP (Garcia-Marlès et al., 2024; Hopke et al., 2022; Trechera et al., 2023; Vörösmarty et al., 2024).

Aircraft engines emit a large fraction of UFP as nucleation mode (NUC hereafter) particles (Brock et al., 2000). Not surprisingly, airports are significant sources of elevated UFP as well, particularly in the NUC range (Lorentz et al., 2019; Stacey, 2019; Rivas et al., 2020). NUC formation after aircraft exhaust involves a dynamic interplay of chemical species. Sulphuric acid often acts as the primary nucleating agent (Schneider, 2005). Lower fuel sulphur content increases the relative contribution of organic compounds to NUC formation (Yu et al., 1999). Jet engine lubricants contribute to the UFP mass fraction (Ungeheuer et al., 2021). Atmospheric observations downwind from airports typically show elevated NUC concentrations most pronounced at about 5–10 km: Los Angeles showed 4-fold increases at 10 km (Hudda et al., 2014). Amsterdam exhibited 3-fold increases at 7 km (Keuken et al., 2015). Boston demonstrated 1.6 to 3-fold increases at 5 km (Hudda et al., 2018). Zurich modelling suggested 2 to 10-fold increases at 3 km (Zhang et al., 2020). London measurements revealed 10-fold increases at 1.2 km (Masiol et al., 2017). At distances exceeding 20 km, tower measurements show a shift towards Aitken mode particles (AIT, 30–100 nm). This shift reflects particle growth from NUC to AIT over several hours (Harrison et al., 2019; Keuken et al., 2015).

Data on atmospheric UFP have been collected over long enough time periods to identify trends. Long-term observations in North America suggest an increase in UFP over time (Chen et al., 2022). In Germany and Europe, however, decreasing trends were observed (Sun et al., 2020; Trechera et al., 2023). This decrease was associated with efforts on air quality control, although these studies did not explicitly investigate the influence of aircraft emissions.

This work presents new PNSD data from an urban background station in the Rhine-Main area of Germany, collected between 2015 and 2021, and builds upon previous research into UFP in the vicinity of FRA (Gregor et al., 2015). Our objective is to examine the impact of anthropogenic sources on local air quality. In particular, we focus on NUC and UFP concentrations from Frankfurt Airport at a distance of 6 km, considering the impact of time of day, season and wind direction. We also assess the specific impact of the lockdown period during COVID-19 pandemic, with its reductions in traffic (Putaud et al., 2021). To our knowledge, this is the first study to examine the impact of an airport on a downwind site over a six-year period."

CR-1: Please always use standardized units and consistent formatting (cm^{-3}, $m\ s^{-1}$)

AC: We revised the script and looked for inconsistent formatting. We therefore use consistent formatting: "number x cm^-3 and m x s^-1 ". Like in table 1 and table 2. Also in line 301 ff " at wind speeds of less than 3 mxs^-1."

line 175: "L min^-1⁻"

line 20, 21 “ (-2%xyear^-1/year)”, “(-5%xyear^-1”, “ (4%xyear^-1)”

line125-128 3 times: “ vehicles x day^-1”

Dear Referee, if it is necessary for this journal we could consistently format the units in the form (cm^{-3}, $m\ s^{-1}$).

CR-1: Usually the size distribution of PNC is divided into two modes: 10–20 nm (nucleation particles) and 20–100 nm (Aitken particles). Why do the authors consider a wider range(10–30 nm) for the nucleation particles? A brief justification for selecting these specific cutoff sizes (10/30/100 nm) would improve clarity.

AC: We have added a sentence in line 190 to clarify this.: “The specific size range for NUC often depends on the instrumentation used for measurement, the specific scientific question being addressed, and the environment being studied (e.g., urban, remote, marine). It is currently defined as 3-25 nm (Wehner et al., 2005) but with higher lower and upper limit for modelling aerosols 10-40 nm (Lupascu et al., 2015). In this study, we defined NUC with a lower limit of 10nm based on the equipment used (Trechera et al., 2023), and an upper limit of 30nm to capture as well the slight overlap of nucleation mode particles with Aitken mode particles.”

CR-1: Although the authors find a correlation with the different emission sources (airport, highways, industrial sources, nucleation) and discuss these aspects qualitatively, a more quantitative source apportionment (e.g., PMF or clustering) would strengthen attribution, especially for the AIT and ACC modes.

AC: We thank the reviewer for this valuable suggestion. We acknowledge that quantitative source apportionment methods such as PMF or clustering would indeed provide additional insights, particularly for the AIT and ACC modes. However, implementing these advanced statistical techniques rigorously would require substantial additional analysis and validation work that extends beyond the scope of the current manuscript and our available resources within the revision timeline.

We believe our current qualitative assessment, supported by meteorological data and temporal patterns, provides a robust first-order qualitative characterisation of the emission sources. To facilitate future quantitative investigations by the scientific community, we have made our complete dataset publicly available (line 488). We will certainly consider implementing PMF or clustering approaches in our future work on a widened dataset adding period 2022-2025.

CR-1: MLH is repeatedly mentioned as a key driver of diurnal and seasonal variability but is not directly examined. If MLH data are available incorporate them into the diurnal and seasonal variability analysis. If not, state limitations more clearly.

AC: Unfortunately we haven't had the possibility to measure the MLH, but we assume from literature that:

Added in line 45 “The MLH in Germany exhibits clear diurnal and annual variations. Generally, the MLH is lower during the night and in the winter. (Ma, 2013; Geiss, 2017).”

Added in line 319: “Studies have shown that wood combustion is a significant source of particulate matter during winter (Pinxteren, 2016).”

Geiß, A., et al., 2017, ‘Mixing layer height as an indicator for urban air quality?’, Atmospheric Measurement Techniques 10(8), pp. 2969-2988 (DOI: 10.5194/amt-10-2969-2017).

Ma, N., et al., 2014, 'Tropospheric aerosol scattering and absorption over central Europe: a closure study for the dry particle state', Atmospheric Chemistry and Physics 14(12), pp. 6241-6259 (DOI: 10.5194/acp-14-6241-2014).

Pinxteren, D. van, et al., 2016, 'Regional air quality in Leipzig, Germany: detailed source apportionment of size-resolved aerosol particles and comparison with the year 2000', Faraday Discussions 189(0), pp. 291-315 (DOI: 10.1039/C5FD00228A).

CR-1: Some specific remarks highlighted with possibly helping input from your side:

Line 59-60: Aircraft emit a large fraction of the UFP in the size of NUC particles. These NUC were primarily caused by VOC emissions"...The sentence is not clear and should be clarified and expanded to better explain the formation mechanisms."

AC: We changed in line 61:"These NUC formed after aircraft exhaust can be attributed to a dynamic interplay. Sulfuric acid often acts as the primary nucleating agent, creating the initial seed particles (Schneider, 2005). The lower the sulfur content of the fuel, the more important the relative contribution of organic compounds becomes to the initial particle formation event (Yu et al. 1999)."

Schneider, J., et al., 2005, 'Nucleation Particles in Diesel Exhaust: Composition Inferred from In Situ Mass Spectrometric Analysis', Environmental Science & Technology 39(16), pp. 6153-6161 (DOI: 10.1021/es049427m).

Yu, F., et al., 1999, 'The possible role of organics in the formation and evolution of ultrafine aircraft particles', Journal of Geophysical Research: Atmospheres 104(D4), pp. 4079-4087 (DOI: 10.1029/1998JD200062).

CR-1: Line 341: "The high midday TNC peak is attributed to regional or urban photo-nucleation..."I suggest "The pronounced midday TNC peak is consistent with regional or urban photochemical nucleation processes...".

Did the authors actually identify the days on which the NPF events occurred? I would suggest isolating the secondary aerosol contribution due to NPF events to better quantify the primary emission contribution from the various sources. The distinction between NPF-related NUC and airport-related NUC could be made sharper

AC: We changed the text in line 341 to:" The pronounced midday TNC peak is consistent with regional or urban photo-nucleation processes, as well as fumigation from higher atmospheric layers rich in nucleation mode particles and ozone. The MLH grows due to convective dynamics, aviation, and/or power plants. This was observed with a higher midday peak in summer, compared to none in winter (see Fig. 8), which was attributed to photo-nucleation."

The authors didn´t identify the NPF events and days respectively. This is beyond the scope of the focus of this article. The approach of the Referee might be a good approach for further evaluations in the future to distinguish between NUC stemming from NPF against primary combustion from various sources.

CR-2: (...) "While most of my comments are minor, my main recommendation is that the authors carefully improve the writing, strengthen the connections between ideas (e.g., the introduction currently consists of short paragraphs with abrupt transitions), and reconsider the organization of some sections, as certain parts could be merged for better coherence."

AC: The authors have revised the introduction to improve coherence and readability. We have also broken up long sentences to improve readability (see AC to CR1).

Dear reviewer, which parts would you suggest to merge beside 3.7?

CR-2: L30 – The first paragraph ends with “we always use PM as an abbreviation for PM mass concentration”. This definition could be introduced earlier, at line 28, where the term “PM” first appears.

AC: moved to line 29: In this manuscript, we always use PM as an abbreviation for particulate matter mass concentration.

CR-2: L32–36 – Consider referencing the new European directive that includes the requirement to measure this metric.

AC: We added in line 38: “… The new EU Air Quality Directive 2024/2881 (Anon, 2024) includes the requirement to measure Particle Number Concentration (PNC) and Particle Size Distribution (PSD). …”

Anon: Directive - EU - 2024/2881 - EN - EUR-Lex, 2024.

CR-2: L38–39 – Clarify that new particle formation (NPF) and nucleation are not identical. NPF includes nucleation, but nucleation can occur without subsequent particle growth.

AC: We added in line 444: ”The combination of nucleation and subsequent particle growth and coagulation into larger particle sizes is usually termed New Particle Formation (NPF).”

CR-2: L95–105 – Explicitly state that the main measurements are PNSD at LAN, while the other stations provide auxiliary data. This key information is currently difficult to locate.

AC: Dear referee, we state this here: line 127 : In addition to the PNSD data from LAN, we also used data from three additional observation sites:

We also mention this in line 99 in the description of fig. 1: LAN provided data on TNC, NUC, AIT, and ACC. DAR, FFM_AP, and LAN_TC provide auxiliary data.

CR-2: L101 – While not critical, I do not see a strong reason to exclude the entire year 2016. It could be omitted from seasonal variation analysis but still used for trend or wind analysis.

AC: We thank the reviewer for this suggestion. The exclusion of 2016 was based on the limited data coverage (~20%) resulting from maintenance work and instrumental modifications during that year. We were concerned that the irregular temporal distribution of these data could introduce bias, particularly in seasonal analyses. However, we recognize that partial data may still be useful for certain applications (e.g., case studies or supplementary trend analysis). For this reason, we have made the complete dataset, including all available 2016 data, publicly accessible via the repository referenced in line 508, enabling other researchers to evaluate these data according to their specific needs.

CR-2: L102 – Remove the word “mass” from “PM mass concentration.”

AC: Dear referee, we removed it. Now at line 111: PM concentration measurements at LAN …

CR-2: L155 – The statement “resulting in larger particle sizes and, as previously noted, an even higher proportion of NUC” is unclear. If larger particles dominate, why would the proportion of NUC increase? Please clarify.

AC: Dear referee, we rephrase the sentence to clarify:

In this instance, hygroscopic characteristics and other water uptake processes may have caused the particles to grow, resulting in larger particle sizes. Smaller particles than 10 nm also grow and would therefore lead to an even higher proportion of NUC.

CR-2: L182 – These quantities are introduced too late, even after their first use in Figure 1's caption. Define NUC earlier. Also, why is NUC defined as 10–30 nm? Typically, 20–25 nm is used as the upper limit. Trechera et al. (2023) use 25 nm, not 30 nm as stated in line 188.

AC: This is also discussed in the answer to CR1 from line 180.

CR-2: Sect. 3.1 – Consider merging this section with another or improving its content. Its title, "Particle number size distributions," is misleading because it does not include PNSDs.

AC: renamed to:" Particle number concentrations of size fractions"

CR-2: Sect. 3.2 – Several figures are presented without clear explanation or connection to the text. Present results sequentially, linking them logically, and introduce figures where they are needed to support the discussion. For example, Figure 2 lacks trend values, which seem important since they suggest extreme episodes are decreasing while others remain stable or show an upward trend (though this is hard to confirm without numbers).

AC: Dear Referee, we rearranged the whole chapter 3.2 and rearranged the figures as well to sequentially discuss the figures and give logical links. Figure 2 displays the temporal trend of seasonal mean TNC at LAN from 2015 to 2021. The regression line shown was computed using R, but the corresponding statistical parameters are not reported. This omission is deliberate to prevent methodological confusion, given that seasonal trends are assessed via linear regression whereas the Theil-Sen robust trend estimator is applied in subsequent figures.

We therefore rephrased the and rearranged the chapter and figures to:

"Figure 2 displays the temporal trend of seasonal mean TNC at LAN from 2015 to 2021, with seasons defined as 3-month periods (winter: December-February).

Figure 2. Trend of seasonal means of TNC from 2015 to 2021 at LAN

The regression line (computed using R) is shown without statistical parameters to maintain methodological clarity, as linear regression is applied here while the Theil-Sen estimator is used in subsequent figures. The moving average reveals a decreasing trend for the 95th percentile, with a pronounced 25% drop in spring 2020

The drop of 25% in 95th percentile in spring 2020 (Fig. 2) corresponded to the COVID-19-lockdown-related decrease in traffic. lLandings and take-offs (LTO) decreased by 70% (Schultheiß-Münch et al., 2022) (s. 3.6) and car traffic on motorways by 16% (BAST, 2023) in 2020 compared to 2019 for car traffic (at LAN_TC) and 2015-2019 for airports. Several studies have examined the environmental impacts of traffic restrictions during the COVID-19 lockdown. In Spain, COVID-19 lockdown restrictions reduced traffic intensity by up to 80%, resulting in lower levels of combustion-related pollutants NO_2, CO, and SO_2 (Putaud et al., 2021; Querol et al., 2021).

Figure S2 shows annual box plots for TNC to complete the picture. The yearly medians confirm the general downward trend.

Figure 3a-c Theil-Sen trend (deseasoned) for monthly means from 2015 to 2021 for all wind directions for (a) LAN: TNC, DAR: (b) PM10, and (c) NO2.

To quantify long-term changes, we applied Theil-Sen trend analysis (see Sect. 2.3) to monthly means of daily values from 2015 to 2021 (Table S6).For better representation, Fig. 3 shows trend lines according to Theil-Sen (s. 2.3) from 2015 to 2021 on the basis of monthly means of daily mean values (cf. Tab. S6). The trend for TNC (Fig. 3a) yielded a highly significant (P<0.001) decrease of -2.3% per annum (p.a.). Even for the shorter period of 2015–2019 when excluding year 2020 as a special year of pandemic-caused reductions, the trend was still -1.9 % p. a. but less significant (P<0.05) (cf. Tab. S6). Trend analyses for an earlier period (2009-2018) at German Ultrafine Aerosol Network (GUAN) stations in Germany showed a stronger decrease between -2.6% to -6.3% p.a. for TNC (20–800 nm) (Sun et al., 2020).

Other components also showed a highly significant (P<0.001) decrease, with even higher rates: PM10 -4.3% p.a. (Fig. 3b) and NO_2 -4.9% p.a. (Fig. 3c). In contrast, some components showed a significant (P<0.01) increase: CO 3.5% p.a., ozone 1.5% p.a. (cf. Tab. S4).

When considering Tab. S4 the decrease for the different particle size ranges was the highest for NUC -2.9% p.a. at P< 0.001 (2015-2021). The decrease was lower for AIT -2.0% p.a. (2015-2021). ACC showed no significant trend or even showed an increase of 3.3% p.a. 2015-2019. Overall, the decrease in TNC seems to be associated primarily with a decrease in the subfraction NUC. This decrease is in line with decreasing trends in other combustion-related compounds of mainly anthropogenic origin, such as NO_2, PM_{10} and cf. Tab. S4 NO and SO_2. In contrast, CO, another combustion-related pollutant, increased as well as ozone.

Figure 4a-c. Theil-Sen trend (deseasoned) for monthly means, 2015-2021, from wind from airport: WR 270°-340°, for (a) LAN: TNC, DAR: (b) PM10 and (c) NO2.

Figure 5a-c. Theil-Sen trend (deseasoned) for monthly means, 2015-2021, from wind not from airport: WR 350°–360° + 0°-260°, for (a) LAN: TNC, DAR: (b) PM10 and (c) NO2.

The airport as an additional source of NUC could be the reason for the lower decline rates compared to the other locations. When only wind from the airport is considered, the trend is higher (-3.7% per year, 2015 to 2021, s. Fig. 4a). Here the pandemic year 2020 shows a dip before increasing again in 2021. This could be due to reduced emissions from the airport. The decrease in wind from the other directions was lower at -1.8% p.a. (Fig. 5a) and showed no visible impact in 2020."

CR-2: Figure 6 – Correct the x-axis label ("year").

AC: Dear referee, we corrected the x-axis label to "2015-2021 (2016 excluded)".

CR-2: L271–272 – Rewrite this sentence for clarity ("lower than"?; is ambiguous).

AC: line 374: Dear referee, we rephrased the sentence to: "In LAN (see Fig. S5), we also observed a small TNC peak during late morning to midday. This midday TNC peak showed lower concentrations than the morning traffic rush hour peak. In contrast, the midday NUC peak exceeded the morning rush hour concentrations. The highest concentrations for both TNC and NUC were found during night hours (21:00-0:00), probably due to nocturnal inversion layers."

CR-2: Figure 7 – Specify what is represented (e.g., particle number concentration; cm^{-3}). The caption is also incorrect ("Polar plot for particles in cm^{-3}").

AC: Dear referee, we rephrased the caption of the fgure to: “Figure 7: Polarplot for particle number concentration in cm^-3, average of hours separated from 2015 to 2021 (excluding 2016), in LAN. (a) NUC, (b) AIT and (c) ACC. Grey spots represent cases less than 2 hours.”

We also change caption fig 6 to: “Figure 6. Polarplot for particle number concentration in cm^-3, average of hours of the years 2015-2021 (2016 excluded),

We also added abbreviation PNC to caption table 1: Table 1. List of air quality sites supplying datasets to this study, with location and type of environment. Coor., coordinates; Alt., altitude; UB, urban background; Met, meteorological parameters; TC, traffic count; PNC, particle number concentration.

and changed caption table headline to “PNC in cm-3”

CR-2: Sect. 3.7 – A dedicated section for limitations may not be necessary. Some limitations could be integrated into the relevant sections, while others might be omitted.

AC: We deleted or moved chapter 3.7:

moved to 2.3: In the LAN data set 2015-2019, high particle number concentrations were detected in the finest size fractions (Trechera et al., 2023). A measurement in the 10 nm range places high demands on instrumental detection, as the CPC used has a low detection efficiency in that range. Therefore, instrument-to-instrument variability has been observed to be higher below 20 nm than above this size (Wiedensohler et al., 2012). However, this higher variability has considerable effects, especially on the number concentrations in NUC.

deleted because information is already included already in 2.1: This study is based on a particle number size distribution (PNSD) dataset from 2015-2021, as well as auxiliary pollutants collected from a federal state authority's air quality monitoring network. It is also worth noting that all datasets had more than 55% data coverage during the study period; one year was excluded due to low data coverage. This shortfall could be attributed to the complexities of UFP-PNSD measurements, as well as the need for close supervision and frequent instrumentation maintenance.

moved to 2.1: The longer distance, particularly between the two stations LAN and DAR, made it difficult to adequately compare the gaseous pollutants to PNSD.

moved to 2.2: We recommend that future studies meet the UFP measurement requirements of ACTRIS and CEN (CEN, 2023, 2020), as well as Wiedensohler et al., 2018, 2012. Therefore, in future we will use an adapted MPSS of TROPOS for PNSD from 10 to 800 nm, including improved sampling according to CEN. Then, we will dry the sampled aerosol below 40% RH and use a PM2.5 inlet as is recommended in the Service Tool 1 prepared within the "Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS" EU-project (RI-URBANS, 2024).