



Analysis of carbon and nitrogen from atmospheric sources by bulk deposition sampling at various locations in Germany

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ABSTRACT

Atmospheric deposition of particulate matter is an important indicator of air pollution and a significant factor in material surface fouling. The elemental composition of this nutrient-containing dust depends largely on the exposure region and time as well as on climate. Therefore, in this paper we report an analysis of atmospheric pollutions with a self-made low-cost bulk deposition sampler directed at sampling deposition via air transport and rainfall. We used the device in diverse environments - thus comparing an urban region, an area surrounded by forest and an area mainly dominated by agriculture. The total organic carbon (TOC) and total nitrogen (TN) amounts were selected as indicator parameters and analyzed in a biweekly rhythm for three and a half and two years, respectively. The TOC value responded to particulate matter in the urban area, especially significant were the influences of the New Year's firework in urban and pollen in the rural forest area. In contrast, the TN value was more under the influence of the nitrogen emissions in the agriculture-dominated area. However, the TN value did not correlate with the NO_x values in the urban area because the atmospheric nitrogen emissions in the city might originate from various emission sources. Summarizing, the TOC and TN values of the self-made low-cost bulk deposition sampler were in good agreement with environmental events of their immediate surrounding. Moreover, the selected containers and sampling procedures are universally applicable to monitor and analyze organic as well as inorganic parameters (e.g. metal ions) of atmospheric deposition.

1. Introduction

Ambient air pollution accounts for stroke, heart disease, lung cancer, acute and chronic respiratory and other diseases of humans (Almetwally et al., 2020; Bo et al., 2021; Januszek et al., 2020; Peled, 2011; VoPham and Jones, 2023). Most of the outdoor pollution sources include residential energy for heating, vehicles, power generation, agriculture, waste incineration, and other industry (Rattanapotanan et al., 2023).

However, air pollution also affects all species of animals and plants (Bazyar et al., 2019) and serves as an important factor in material surface fouling (e.g. effect of dust composition on the reversibility of photovoltaic panel soiling (Varga and Wiesner, 2021)). The dust that can cause material surfaces to be colonized by microorganisms can have various sources e.g. bird droppings, pollen, agricultural emissions, engine exhaust and mineral dust (Kazem, 2023). Carbon and nitrogen emissions in particular are a suitable breeding ground for the growth of microorganism (Ramos et al., 2023).

Previous studies (Decina et al., 2018; Zhang et al., 2021) showed that wet and dry deposition sampling is a useful technique to analyze how carbon and nitrogen deposition on the earth's surface has adverse impacts on ecosystems and humans. This way environment changes can be detected and analyzed which serve as an important indicator for air pollution as well as detect nutrient inputs that play a role in material surface fouling. Therefore, the aim of the paper was to analyze and monitor annual fluctuations in carbon and nitrogen atmospheric depositions in three different geographical regions.

The experimental design was based on passive sampling of atmospheric deposition because grab or composite samples provide only a snapshot of the current state and the overall amount of the pollutants, especially over longer time intervals, could be much higher. Passive sampling is based on the free flow of analyte molecules from the sampled medium to a collecting medium. For total (wet and dry i.e. bulk) deposition sampling of atmospheric deposition various devices were used previously (Erisman et al., 1994; Khuriganova, 2019; Nežiková

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et al., 2019; Pan et al., 2021; Waza et al., 2019).

For sampling of the atmospheric depositions, we designed a simple bulk deposition sampler for carbon and nitrogen atmospheric deposition. The potential of our low-cost bulk deposition sampler was used in a long-term monitoring of atmospheric deposition in three different locations: an urban region of Berlin, an area surrounded with forest in Horstwalde, state of Brandenburg and an area mainly dominated by agriculture in Neufahrn, Bavaria.

2. Materials and methods

2.1. Locations and bulk deposition sampling

The bulk deposition sampling devices were used for dry and wet deposition samples and were installed on three different locations: A urban location at BAM headquarter in Berlin-Lichterfelde, Germany, a forest location at the BAM Test Site Technical Safety in Horstwalde, state of Brandenburg, Germany and a mainly agricultural dominated location in Neufahrn, Lower Bavaria, Germany (Fig. 1 shows the three locations in Germany).

The urban location (coordinates: N 52.441704, O13.286133) is on the campus of the BAM headquarter which is located between the multi-line entrance road "Unter den Eichen" and a railroad line in the south-west of Berlin, Germany. The forest location is on the BAM test site in Horstwalde (coordinates: N 52.095556, O 13.407435) which is far from any city or village surrounded by a large pine forest. The agricultural dominated location (coordinates: N 48.722662, O 12.185149) is on the plant site of Erlus AG in Neufahrn, Lower Bavaria, Germany which is surrounded by agricultural fields. The experiments run from beginning 2017 to mid-2020 at the locations Berlin and Horstwalde and from mid-2018 to mid-2020 in Neufahrn, respectively.

The developed bulk deposition sampler consists of a round food-safe HDPE plastic container with a volume of 3.7 L (see Fig. 2). To avoid the impact of large particles, this container is covered by a polymer insect grid with a diameter of 110 mm and a grid size of $1 \times 1 \text{ mm}^2$. The bulk deposition sampler was placed at all three locations at the end of a roof (see Fig. 2). On all locations two devices were used for sampling of carbon and nitrogen and an additional device was closed with a lid as a blank reference. The results of the two devices at each location are shown hereafter as TOC1/2 and TN1/2. 500 mL of bi-distilled water was



Fig. 1. Bulk deposition sampling locations in Germany



Fig. 2. The used low-cost bulk deposition sampler applied on a roof

added (due to the higher loss caused by evaporation between May and September 1000 mL were used) into each bulk deposition sampler device. Every 14 days the content was sampled, the volume of the solutions determined and the concentration of total organic carbon (TOC) and total nitrogen (TN) in the solutions analyzed. The plastic containers were cleaned and re-used every time.

2.2. Total organic carbon (TOC) and Total nitrogen (TN) analysis

The concentration of dissolved TOC and TN in the aqueous solutions from the bulk deposition sampler were analyzed after filtration through a glass microfibre filter (GF 6, Size: 55mm, weight: 80g/m², Whatman, Maidstone, UK) with the Shimadzu TOC-V_{CDH/CPN} instrument (Kyoto, Japan). The TOC and TN values were specified in mg/L. Afterwards, this value was multiplied with the final volume of the aqueous solutions from the bulk deposition sampler to determine the total amount of TOC and TN in mg for the sampling period of two weeks. Finally, the values are divided by the area of the sampler and expressed in $\mu\text{g}/\text{m}^2$.

2.3. Meteorological/weather data

The bulk deposition sampling stations in Berlin and Horstwalde are only a few meters away from a meteorological station of Deutscher Wetterdienst (DWD). The Figs. 3 and 4 show the monthly average temperature and the monthly cumulative rainfall and sunshine duration, respectively of these locations. Because the station in Berlin-Lichterfelde did not count the sunshine duration the data from the DWD station in Berlin-Tempelhof were used. Additionally, the wind directions and wind speeds of these locations were continuously collected by DWD. For the location Neufahrn the wind directions and wind speeds were used from the closest DWD station at Munich airport.

2.4. Particulate matter and NO_x data

The particulate matter (PM₁₀ = particulate matter with an aerodynamic size of $< 10 \mu\text{m}$) and NO_x data from the air pollution monitoring station Schildhornstraße in Berlin-Steglitz were downloaded from the website: <https://luftdaten.berlin.de/station/mc117>. This station is the closest station to our bulk deposition sampling station in Berlin-Lichterfelde (approx. 3 km) and has also a very similar traffic volume. The data are displayed as monthly average in Fig. 3.

3. Results and discussion

3.1. Carbon and nitrogen atmospheric deposition on the various locations

Figs. 3–5 top show the collected amounts of TOC and TN from the

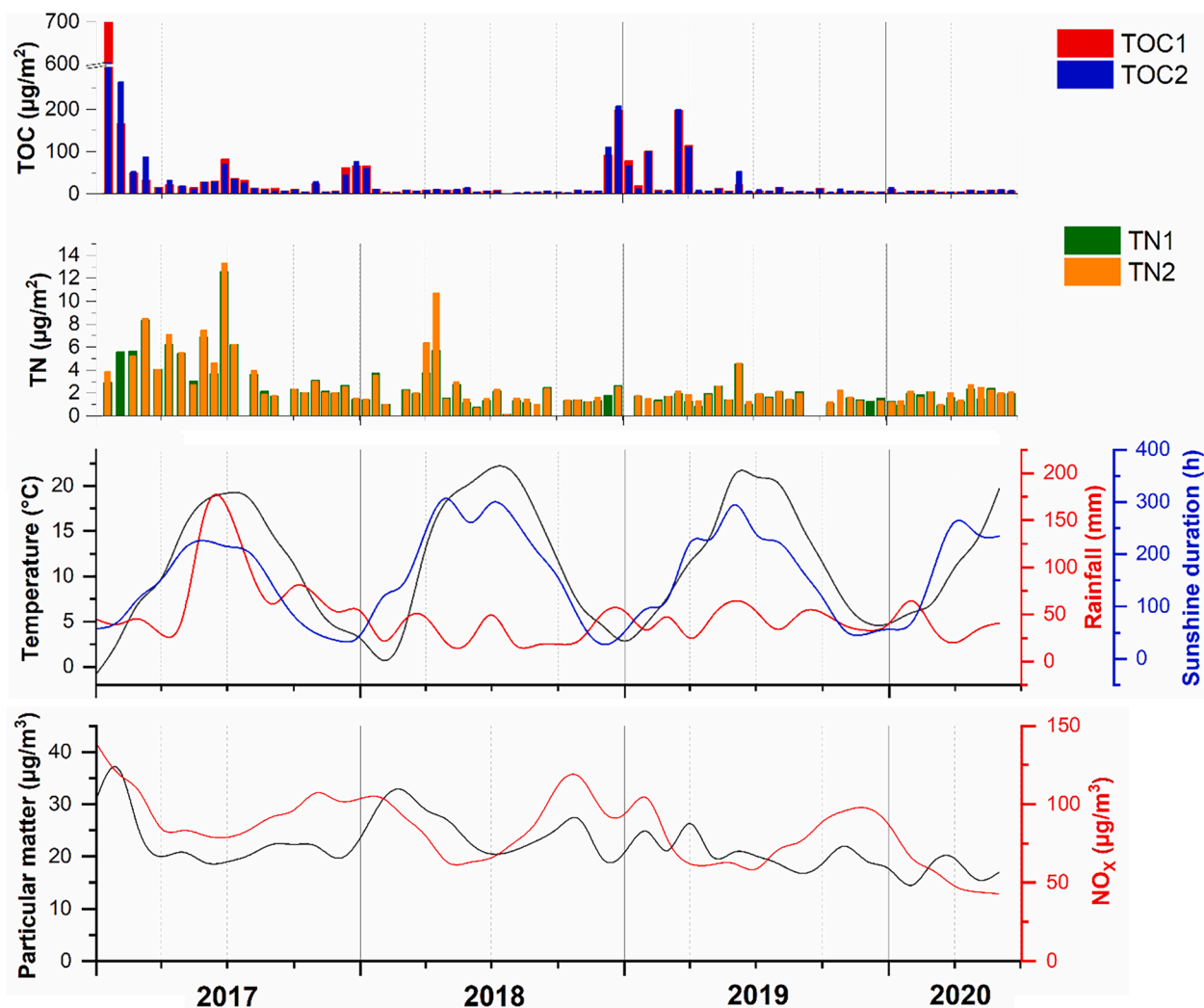


Fig. 3. Amount of TOC and TN (in $\mu\text{g}/\text{m}^2$) of the two bulk deposition sampler (TOC1/2 and TN1/2) from beginning 2017 to mid-2020 (top) and metrological data (middle) from the location Berlin-Lichterfelde; Particulate matter and NO_x data from the location Berlin-Steglitz (approx. 3 km distance; bottom)

low-cost bulk deposition sampler over the period from beginning 2017 to mid-2020 and mid-2018 to mid-2020, respectively, for the three locations. The data from all three stations show differences in TOC and TN values during varied periods of the year which will be discussed in more detail in the following sections.

The urban location Berlin-Lichterfelde shows, because of the high amount of fine dust in the city, generally a higher level of TOC (almost a magnitude) than the other two locations which always reaches its maximum in the beginning of the year. It reached the maximum for the beginning of the year 2017 and decreases for the beginning of the years 2018, 2019 and 2020. Additionally, the particulate matter at the air monitoring station in Berlin-Steglitz, close to our bulk deposition sampling station in Berlin-Lichterfelde, correlates nicely with the TOC data (see Fig. 3 bottom). Other researchers showed that the New Year firework has a strong impact on particulate matter in ambient air (Hoyos et al., 2019; Lin, 2016). Thus, the bulk deposition sampler collects the particulate matter from the seasonal firework which was displayed at the beginning of the new year. Dauert et al. (2021) showed that the New Year firework is the highest particulate matter pollution of the year in Germany. Furthermore, the authors also showed that there was a strong decrease of particulate matter at New Year 2018 in comparison to New Year 2017 and again a slightly increase at New Year 2019. This agrees also with our data from the bulk deposition sampler.

The station in Horstwalde, which is located surrounded in a forest in

the state of Brandenburg (see location in Figs. 1 and 7), show another phenomenon. Here, every year increased amounts of TOC and TN were detected in the period from April to July (Fig. 4 top). This agrees with the pollen season of pine trees (Polleninformationsdienst, 2020), which is the major tree species in the forest around Horstwalde. Thus, the bulk deposition sampler collects pollen which enters the device via air transport and rainfall thus helping us to judge this input into PM deposition.

In general, weather seems to strongly affect the TOC and TN values. The year 2018 was the driest and hottest year in Germany since the weather record began (see also Fig. 4 bottom). This results in much lower TOC and TN values for the location Horstwalde (Fig. 4 top). On one hand there might be a reduced pollen flight in this year because of water stress for the trees. On the other hand, the long dry season without rain might not “washed out” the pollen from the air which enters the bulk deposition sampler subsequently. The influence of the rain was also detected on another phenomenon in May 2017 in Berlin and Horstwalde. In this month there was an unusual high amount of rainfall (see Fig. 3 middle and Fig. 4 bottom). Thus, also here most probably pollen in the air and on rain-exposed surfaces were “washed out” and collected in the bulk deposition sampler which possibly results in higher amounts of TOC and TN than the weeks before and after.

Mostly a similar TOC/TN ratio was found in the bulk deposition sampler. However, at the location Neufahrn in spring 2019 and 2020 there are increased TN values (in comparison to TOC; see Fig. 5).

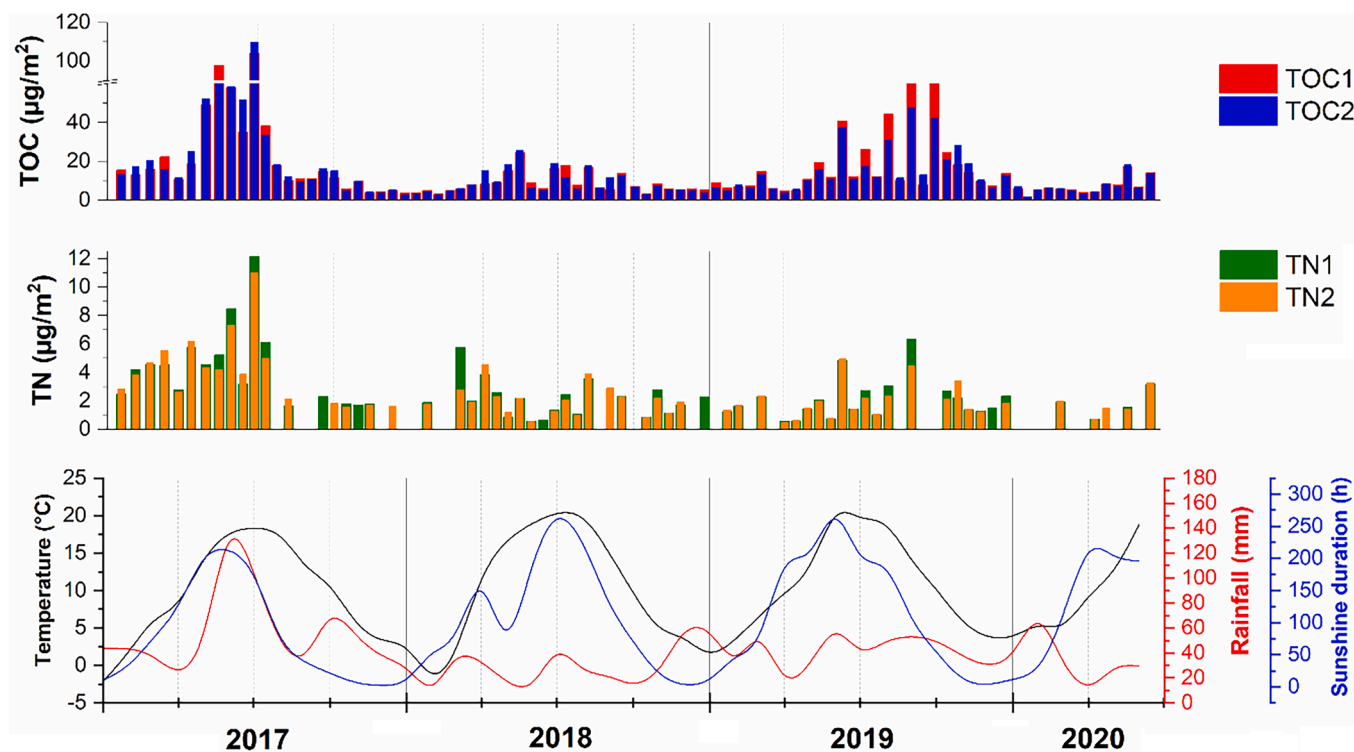


Fig. 4. Amount of TOC and TN (in $\mu\text{g}/\text{m}^2$) of the two bulk deposition sampler (TOC1/2 and TN1/2) from beginning 2017 to mid-2020 (top) and metrological data (bottom) from the location Horstwalde, state of Brandenburg (south of Berlin)

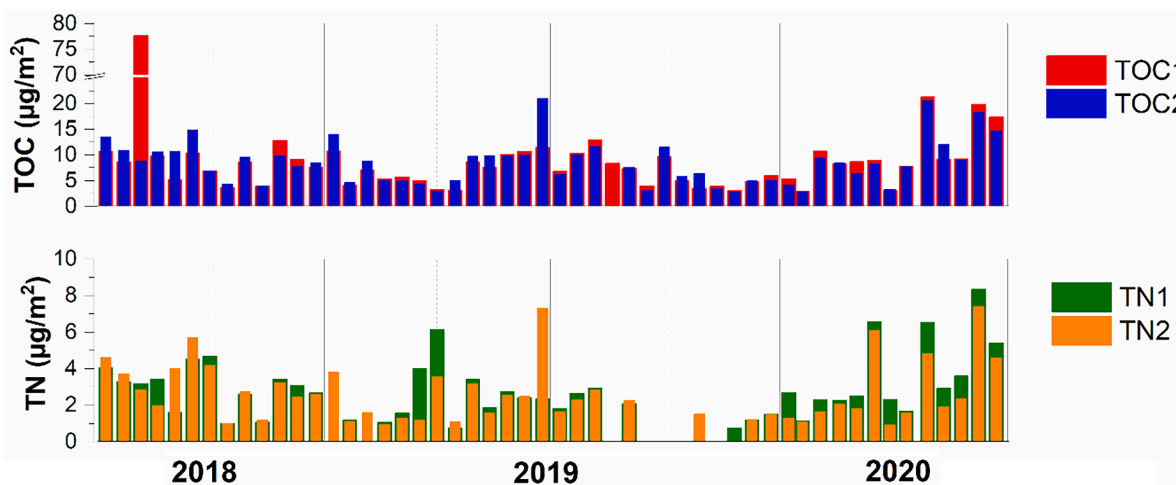


Fig. 5. Amount of TOC and TN (in $\mu\text{g}/\text{m}^2$) of the two bulk deposition sampler (TOC1/2 and TN1/2) from mid-2018 to mid-2020 from the location Neufahrn, Bavaria.

Neufahrn is a predominately agricultural area, where farms are around, especially in the west which is also the main wind direction in that area (see green dots in Fig. 8). There, manure and nitrogen fertilizers were applied to fields in spring which is most probably detectable with the bulk deposition sampler. The negative effect of fertilization with nitrogen compounds is an increased emission of NH_3 and N_2O gas from the amended soils (Saha et al., 2019; Zhou et al., 2017). Yao and Zhang (2013) used also bulk deposition sampler to analyze the emission of nitrogen from agricultural farmland. They showed that the highest nitrogen values were inside the intensive livestock production zones. Thus, most probably the agricultural areas around Neufahrn lead to the higher TN values in spring.

Additionally, other researchers (Bhattarai et al., 2019; Han et al., 2020) showed that biomass burning emissions can have a high impact on

atmospheric nitrogen concentration, which could be also an explanation in the rural area of Neufahrn. Furthermore, the bulk deposition samplers were located on the area of the company Erlus AG in Neufahrn. For their production of roof tiles also natural gas was combusted, whereby also nitrogen compounds occur (Xue et al., 2017). However, there is a continuously production of roof tiles at this plant, thus, these emissions might have a minor effect on the collected data.

3.2. Extraordinary phenomena of atmospheric deposition

The phenomenon of the increased TN values was also detected in spring 2018 at the location Berlin (see Fig. 3). One explanation could be, that normally the wind in Berlin comes from the west/south-west (Fig. 6), but a view on the wind directions in spring 2018 (not shown)

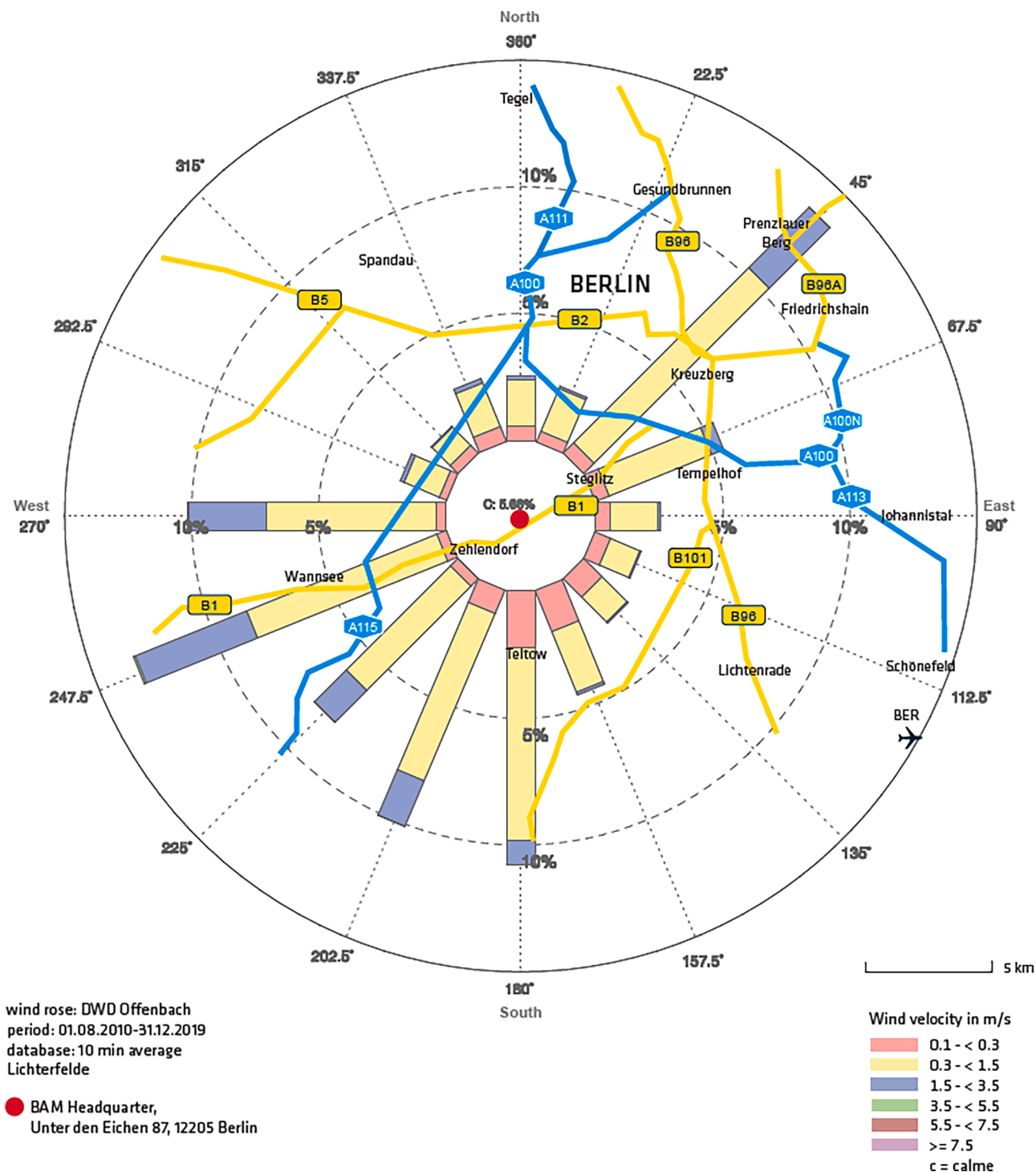


Fig. 6. Average wind rose of the location Berlin-Lichterfelde for the years 2010 to 2019 and sketch of the surrounding landscape.

showed that in that time the wind was mainly from the south/south-west where are agricultural areas in the state of Brandenburg (the province around Berlin).

Thus, also here NH₃/N₂O gas emission from manure/fertilizer application could be the reason of the increased TN values. On the other hand, also here the increased nitrogen content could be originated from biomass burning (Bhattarai et al., 2019; Han et al., 2020) in the province Brandenburg which is typically for this Easter season. However, Han et al. (2020) showed also that in urban areas the nitrogen emissions can have various other emission sources e.g. automobile exhaust. This agrees with our results, because the investigated bulk

deposition sampler possibly collects all kind of atmospheric nitrogen and therefore also no correlation was detectable between the passive sampling TN value and the NO_x concentration from the air pollution monitoring station in Berlin-Steglitz (see Fig. 3). Thus, the phenomenology of the TN atmospheric deposition is so complex that asserting TN peaks cannot be answered conclusively.

Moreover, in autumn 2019 the location Horstwalde shows an unusual high TOC concentration in comparison to TN. Normally, the wind in this location comes from the west/south-west in Horstwalde (see Fig. 7). However, in October 2019 there was a longer period with wind from the east/south-east (data not shown). In direction south-east of

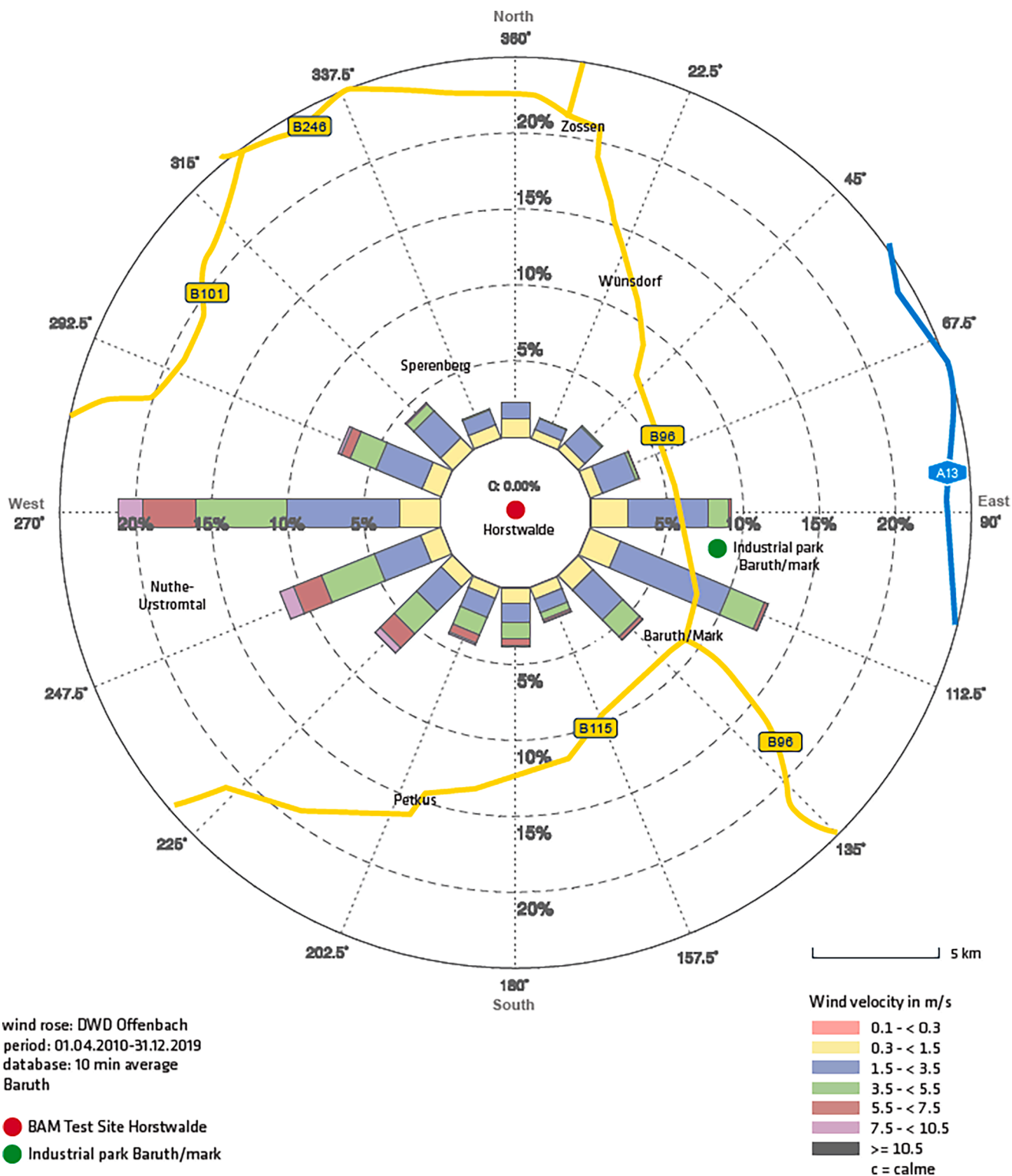


Fig. 7. Average wind rose of the location Horstwalde for the years 2010 to 2019 and sketch of the surrounding landscape. The green dot shows the location of the woodworking industry in Baruth.

Horstwalde is the small city Baruth/Mark (approx. 7 km; see green dot in Fig. 7) where are several companies located from the woodworking industry. Thus, it might be possible that the increased TOC values in that time period come from an entry of sawdust, but there might be also another reason which cannot be unraveled by bulk deposition sampler only.

3.3. Performance of the self-made bulk deposition sampler

Double determinations were always made with two passive sampling collectors per measuring interval (TOC1/2 and TN1/2). This along with a high volume of sampling liquid has the advantage that a conspicuous and highly elevated value for TOC and TN in only one of the two sample collectors is an indication of contamination. This was detected several

water. However, they did not dissolve in the water and therefore their TOC and TN input can be neglected.

4. Conclusions

In our paper, we showed the potential for our new low-cost bulk deposition sampler on sampling of TOC and TN from atmospheric deposition. This long-term sampling study showed that changes of the TOC and TN values could mostly associated with an event in the immediate surrounding. The three different locations – urban, forest and agricultural – showed that the developed bulk deposition sampler is useable for various environmental conditions. While the TOC value was dominant from the high amount of particular matter in the city the TN value correspond to nitrogen emissions in agriculture dominated area. Moreover, the TN value did not correlate with the NO_x values from the air monitoring station in the city, because the atmospheric nitrogen emissions in the city can be from various emission sources. Due to the remote location of Horstwalde in the state of Brandenburg, only the influence of pollen was detectable here. Summarized, the investigated self-made bulk deposition sampler is an economic alternative for long-term monitoring of carbon and nitrogen from atmospheric deposition but also other organic as well as inorganic compounds (e.g. metal ions) can be monitored with it.

CRedit authorship contribution statement

Christian Vogel: Writing – original draft, Visualization, Methodology, Formal analysis. **Klaus Urban:** Writing – review & editing, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Carsten Ackerhans:** Writing – review & editing, Methodology, Investigation, Formal analysis, Conceptualization. **Anna A. Gorbushina:** Writing – review & editing, Supervision, Investigation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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