Reply to RC2

We thank the referee for the review of our manuscript and the valuable comments and suggestions. In the following the referee's comments are repeated (in black) along with our replies (in blue) and changes made to the text in the revised manuscript (in red). Page and line numbers refer to those in the preprint version.

General comments:

The authors present the results of a comprehensive study on the differentiated assessment of inland shipping emissions on the Upper Rhine near Worms in Germany. They use a wide range of measurement techniques to detect gaseous (CO_2, NO_x and O_3) and particulate (PNSD, PNC, PM_x, soot) air pollutants. Two sites have been selected for the measurements, allowing different scenarios to be mapped. One site was located on a bridge in order to record the plumes from passing ships close to the source. The second site was chosen directly on the banks of the Rhine. In this way, it is possible to determine the level of emissions that could affect people living near the Rhine. Particularly noteworthy is the methodology developed to identify individual ship plumes. The algorithm used avoids overlapping plumes, which can be caused by several ships passing at the same time. As a result, only clearly identifiable ship plumes are included in the evaluation. This results in a significantly reduced number of evaluable ship plumes and also reduces the number of individual ships in the composition of the shipping fleet. At the same time, the quality of the subsequent allocation and classification is significantly improved. In particular, the continuous long-term measurements over a period of one year provide a good picture of the emissions of the shipping fleet in this part of the Rhine. In addition, the emission factors can be calculated under real conditions, leading to a better understanding of the impact on inland navigation. This work represents a solid contribution and, in part, a new scientific approach to the measurement and characterisation of emissions from inland navigation under real conditions. The work is recommended for publication by this reviewer. The following suggestions may be incorporated into the authors' opinion.

We thank the referee for the positive evaluation of our manuscript and the helpful comments and suggestions below.

Specific comments:

P3 L21

…high temporal resolution of ~1 s…

Maybe one can mention, that the SMPS has a different and longer temporal resolution for a whole scan of the size range. Additionally, one could also explain the "problem" with scanning devices as a SMPS with a moderate sampling time. The assumption with a scanning device as the SMPS is that the aerosol spectrum does not change much over the time of a scan. However, this can occur with passing ships and short-term increases and thus lead to a distorted PNSD.

We agree that this is an important point to mention and added the following sentences to the text.

An essential part was the application of a fast mobility particle sizer (FMPS) measuring the whole size distribution in parallel by the use of numerous electrometers. For the detection of short-term increases like ship plumes it has a crucial advantage over commonly used scanning mobility particle sizers (SMPS) which need several tens of seconds for a scan and thus depend on the size spectrum not changing significantly over time.

P4 L11

…Instrument-specific sampling lines of 4-5 m length…

It seems that the calculated particle loss under 10 percent is relatively low. I would expect a higher particle penetration at this length of the sampling line. Did you use separeted sampling lines or did you use one sampling line with a higher volume flow and a manifold leading to the individual measuring devices?

We used separate sampling lines for each instrument at BRI but the flows for FMPS and AE33 were relatively high (10 lpm and 5 lpm respectively) so that diffusion losses for small particles could be limited. From Fig. S2 in the supplement it can be concluded that for the Grimm 11-D device (1.2 lpm flow rate) the overall loss rate for particles with diameter < 2.5 µm remains below 10 % but increases to more than 50 % at 10 µm. This is why we compared the measurement at BRI with the optical particle counter at RIV to make sure that the low particle count rate at diameters > 2.5 µm is a real feature of shipping emissions and not an artefact of the potentially high losses in the sampling line at BRI. We added the flow rates of the three devices to Fig. S2.

Calculated overall transmission losses with ~ 5 m sampling line for the instruments FMPS (flow rate 10 lpm), Grimm 11-D (flow rate 1.2 lpm) and AE33 (flow rate 5 lpm), derived using the Particle Loss Calculator Tool (von der Weiden et al., 2009).

P4 L12

…to enable an undisturbed incoming flow.

Doesn't the bridge itself generate turbulence that can contribute to influencing the wind field at the measurement site? Are downwind eddies possible that carry road traffic emissions down to the measurement site and superimpose the ship plumes as well?

We agree that the word ‘undisturbed’ is misleading. We tried to realize an incoming flow as undisturbed as possible within the technical possibilities at the bridge but it is likely that turbulence was generated close to the walls. Downwind eddies from road traffic are theoretically possible but the distance to the sampling line is > 5 m and the signal would clearly differ from the typical peak shape of ship plumes. Occasionally we observed a high atmospheric variability in the NO_x signal (depending on meteorological conditions) which had to come from a local source close by, i.e. probably road traffic. But these periods were excluded in the analysis (see peak finding algorithm criteria) in order to avoid interference from non-ship sources. We modified the text accordingly.

Instrument-specific sampling lines of 4–5 m length were led through a hole in the wall, downwards to a point sharply below the edge of the bridge's base to reduce the impact of turbulence on the incoming flow. This way, the distance to the traffic lane on the bridge was also increased to ~6 m so that the impact of traffic emissions on the measurement site was minimized.

P5 L5

…to avoid strong interferences from road traffic.

You have chosen the locations to also avoid the influence of traffic related air pollutants. I am not familiar with the local conditions, but a look at the Nibelungen Bridge shows that this is a double bridge with two lanes each. What traffic volume can be expected there? Is there rush hour and congestion with traffic jams on the bridge? Especially with winds from northern directions, lee vortices could transport the TRAPs to the sampling point.

Both lanes of the bridge are highly frequented so there is the possibility of traffic jams and increased emissions during rush hour. As explained above, usually we did not notice any influence from road traffic on the bridge above. Occasionally we experienced an increased signal variability that can probably be attributed to road traffic (either emissions transported downwards via turbulent eddies or accumulated during the occurrence of thermal inversions) but this did not bias the analysis (see comment above).

Occasionally we observed an increased atmospheric variability in the NO_x signal that probably originated from local non-ship sources like road traffic (favored in the presence of thermal inversions). These periods were, however, excluded from the analysis by defining appropriate criteria in the peak finding algorithm (see Sect. 2.3.1), so that the results were not biased.

P6 Table 1

Here the temporal resolution from the AIS signals is 1 s. To the best of my knowledge, an inland vessel sends a data set only every 10 s, depending on the current movement status.

Yes, like stated on p.8, l.6, AIS data is usually transmitted every 10 s. We changed the entry in Table 1 to 10 s.

P12 L20-21

…further results […] refer to this instrument.

This sentence is somewhat confusing, since in the coming chapters the results on RIV site will also be reported, which, however, were measured with the SMPS.

Our intention was to point out that - since the FMPS covered > 99 % of all particles detected in ship plumes - the results from the optical particle counter did not have a measurable impact on calculated emission factors and the additional contribution at

BRI. The SMPS measurement at RIV was only used to derive the background signal at RIV, whereas the additional contribution from shipping was estimated from the measured FMPS contribution at BRI (assuming a similar dilution factor as for CO_2). We slightly adapted the statement for clarification.

Since the FMPS (size range 6–520 nm) covered on average > 99.9 % of all particles detected in ship plumes, further results of PNC, PM_1 and $PM_{2.5}$ at BRI were based on this instrument.

P12 L26-27

The study by Pohl et al. was performed in Duesseldorf. So please change Upper to the Lower Rhine.

Thank you for noticing.

These results are consistent with a measurement study by Pohl et al. (2017) at the Lower Rhine.

P14 L16

…as well as modern ships with exhaust after treatment…

With regard to the CLINSH project. Weren't up to 40 ships retrofitted with downstream exhaust aftertreatment systems? Are the data or names of the ships available the authors to specifically read them out in their data set in order to be able to better scale up the positive effect of the emission reduction? This would be a good contribution, especially in view of the continuing increase in shipping traffic in the future.

Indeed, this would have been a good opportunity but from the best of our knowledge none of these ships passed the station in Worms during the measurement period. The majority of the ships retrofitted with exhaust aftertreatment systems in the CLINSH project are operated in the Netherlands or on the Lower Rhine in Germany. Nevertheless, we captured three ships in Worms fulfilling the recent Euro V standard (and thus using exhaust gas aftertreatment) which gave us the possibility to scale up the positive effect of an emission reduction, as also suggested by referee #1 (see our answer to referee #1).

In addition, on p.17, l.3 we now compare the mean emission factor of Euro V ships from this study with the mean emission factor and the emission reduction relative to CCNR II ships reported by the CLINSH project.

This low value and the ~ 90 % NO_x reduction compared to CCNR II vessels agree very well with observations made by CLINSH (2022).

P21 L10

With a BC fraction of 38 % for...

It is (for me) not clear to which correlation the value is. Can you please more specify this. Is it BC_{880} nm to total BC?

This value (measured at 880 nm by the AE33) refers to the fraction of BC mass relative to total PM derived from the FMPS measurement.

With a BC fraction (relative to total PM) of 38 % for upstream ships and 16 % for downstream ships our results indicate that a higher engine load leads to an increase in BC emission.

P21 L14

The proportion coming from biomass burning is mentioned here as about 10 % from biofuel combustion. Could it be a possible reason that the analyzed probe isn't just from ships because you also measure the background were also particles coming from wood fires, cigarette smoke, etc. Maybe there could be a hint, if the amount of bb is higher during the wintertime due to fireplaces?

We analyzed the biomass burning fraction at the time of occurrence of the peak maximum (average peak height ~ 50 µg m^{-3}) so that the background concentration (which was usually below 1 µg m^{-3}) can be considered negligible and a significant bias from non-ship sources can be excluded. We also found no difference between summer and winter season, indicative for residential heating.

The algorithm used by the AE33 to internally calculate the biomass burning fraction is based on multi-wavelength analysis (near-IR absorption relates to BC from combustion; a stronger absorption in the ultra-violet regime relates to organic material typical for biomass burning). Since the signature varies depending on combustion conditions and material, a clear separation can be difficult. Our statement on p.21, l.14 regarding biofuels is quite speculative but there are some hints in the literature that the fuel type has an impact on black / brown carbon measured by the aethalometer, e.g.:

Ma, M., Rivellini, L. H., Kasthuriarachchi, N., Zhu, Q., Zong, Y., Yu, W., Yang, W., Kraft, M., and Lee, A. K.: Effects of polyoxymethylene dimethyl ether (PODEn) blended fuel on diesel engine emission: Insight from soot-particle aerosol mass spectrometry and aethalometer measurements, Atmospheric Environment: X, 18, 100216, https://doi.org/10.1016/j.aeaoa.2023.100216, 2023.

We now write:

From multi-wavelength analysis of the AE33 (Sandradewi et al., 2008) we derived a mean fractional contribution from biomass burning of ~10 %, indicative for organic aerosol components that might be formed during the combustion of ship diesel blended with biofuel.

P21 L25

…(see methods).

Please refer to the chapter.

We now write (see Sect. 2.3.4).