acp-2020-1086 (Walden et al.)
Measurement report: Characterization of uncertainties of fluxes and fuel sulfur content from ship emissions at the Baltic Sea

Reply to Reviewers

We thank the reviewers for their critical, valuable and constructive comments. We have made a major revision and replied to all the comments. Our answers below are written by blue color. The changes made in the manuscript are written below by red color, and highlighted in the manuscript by yellow color. The manuscript was significantly improved.

Referee 1

Walden et al use a gradient method to investigate sea-atmosphere fluxes of various species. The detection limit of the gradient methd is not sufficient to observe exchange fluxes for most gases. The authors report particle deposition fluxes, likely originating from ship emissions. Additionally FSC is assessed. The FSC in my opinion is the most interesting part of the manuscript, why weren't concomitant NOx and particle plumes tracked, as this would seem a natural extension of the experiment.

The emissions factors for NO, NO_x, SO_2, Ntot, and $PM_{2.5}$ as well as the FSCs for the same ships were already estimated in our previous publication Pirjola et al., Atmos. Meas. Tech. 7, 249-261, 2014. Discussion of these results were added in the text.

p. 16, lines 475-479: Also the obtained FSCs were in good agreement with the information given by the ship owners, as well with our earlier results in Pirjola et al. (2014), in which the emissions from the same ships were studied in winter and summer campaigns in 2010 and 2011. The mobile laboratory Sniffer was standing at the harbour areas in Helsinki and Turku. Besides the FSCs we also estimated the emissions factors for NO, NO_x, SO_2, Ntot and $PM_{2.5}$, For example, the emission factors for NOx were in the range of 56 to 100 g $(kg\ fuel)^{-1}$.

As for the flux analysis a number of major issues arise when reading the paper. While presenting CFD calculations to support the measurement site setup, it is not clear whether stationarity criteria were fulfilled due to passing ships and associated plumes being advected over the site. I disagree that stationarity is primarily characterized by concentration trends, which are part of the longer wave spectrum, often filtered out by turbulence averaging intervals. There are better ways to investigate stationarity (see standard textbooks on micrometeorological data pre-processing). As such the interpretation of fluxes needs to be evaluated carefully, because many fundamental criteria often implied for flux measurements might not be fulfilled.

We considered the stationarity as between the flux estimate when the trend in the wind speed was removed and when this trend was not removed. For comparison as a criteria for stationary we used the method proposed by Foken and Wichura (1996) for fluxes of CO_2, heat, water vapor and for momentum. We evaluated the observed fluxes following the QA/QC procedures presented later in the text. We replaced the stationary criteria according to the method of Foken and Wichura into the manuscript. Fundamental criteria for flux measurements stationarity of the flux measurements, the footprint area and the occurrence of swell were considered. The following text was added:

p.3, lines 68-70. The use of micrometeorologocal methods reguires criterias to fulfil for the atmospheric conditions being similar for both methods. As such stationarity of the flux measurements, the footprint area and the occurrence of swell were considered.

p. 5, lines 121 – 124. General conditions for the M-O similarity theory are horizontally homogeneous surface structure, stationary (or near stationary) condition (e.g. Foken and Wichura, 1996), constant flux layer and that the atmospheric turbulence is affecting on the vertical profiles of wind speed, potential temperature and humidity.

This might also relate to different footprints of individual levels of the gradient tower (e.g. is the lowest level even seeing the water surface or partially also influenced by the island?)

We calculated the footprint area at each of the measured height at stable, neutral and at unstable conditions according to Högström et al (2008). Based on the calculation in neutral conditions the footprint area from the lowest height accounts 0.3 % from the observed flux at the distance of 20 m from the mast (i.e. at sea line, see in Fig. 2b) while at altitude of 10.7 m the footprint area starts at 40 m from the mast. In Fig. 8b the distance where the maximum flux (red color area) is gained is presented for different heights.

We added the missing text for the analysis of the footprint area to the final manuscript.

p. 13, lines 379 - 385: The footprint area i.e. the area along the upwind where the exchange of gases and particles between the air-sea surface are expected to be a source of the measurement results, was calculated according to Högström et al. (2008). The footprint area was calculated at each of the measured height at stable, neutral and unstable conditions. Fig. 8a illustrates the relative intensity of the footprint areas in neutral conditions as a function of upwind distance from the measurement mast at instrument heights of 4.7 m, 7.2 m and 10.7 m. The cumulative relative contribution (Fig. 8b) indicates that at the lowest height (4.7 m) less than 0.3 % of the observed flux takes place at the distance of 20 m from the mast reaching 90 % at a distance of 3 km. At the height of 10.7 m, the footprint area starts at 40 m from the mast reaching 85 % at distance of 3 km.

The fact that ship plumes on the order of a few seconds were observed suggests that homogeneity and stationarity was largely not fulfilled for quantifying fluxes from ships.

The open sea sector that is studied here more intensive is to direction where the ships are approaching to or moving away from Helsinki. This means that we measure the ship plumes over the footprint area. The duration of the ship plumes varied from 3 to 7 min. The time is of the same order as the short time averages used in the stationary test by Foken and Wichura (1996). We analyzed single emission peaks from the ships. In Fig 10 is a schematic presentation on the dispersion of ship plume (a) and the momentary plumes at different time intervals (b). In Fig. 11 d and e we present the analyzed ship peaks and compared them with the 30 min average values for CO_2 and N_{tot}.

A comparison between CO2 eddy covariance fluxes and gradient measurements is shown, but it is not indicated what QAQC criteria (e.g. u*, ICT, stationarity) were used to filter data, and how much of the original data was used for the analysis after QAQC filtering. Were storage fluxes considered?

The storage fluxes were not considered at this campaign. The site was at the sea where most of the time the turbulent mixing was the driving force for gas and particle dispersion. The following QA/QC procedures and criteria for calculated fluxes were added in the text:

p. 12, lines 360-377

Quality control (QC) and quality assurance (QA) procedures are actions that should take into account in order to improve the data quality and to make the data comparable with the similar data from other studies. Although QA and QC procedures have slightly different meanings, in this study, the quality assurance and quality control (QA/QC) procedures are considered together. The following QA/QC procedures and criteria for flux calculations were taken into account.

1. Calibration of the used analyzers for gases and particles (Section 3.3), for the GR and EC methods
2. Criteria for minimum concentration difference between the measurement heights (Fig. S2), for the GR method
3. Correction of the wind flow field around the measurement mast according to the CFD calculations (Section 4.1), for the GR method
4. Restriction to open sea, i.e. wind direction in the range of 150-270 degrees (Fig. 3a), for the GR and EC methods
5. Analysis of swell to demonstrate the validity of M-O-theory with Codes 1-3 (Section 4.1), for the GR method
6. Footprint area (homogeneous fetch area) was estimated at each of the measurement height and at neutral, stable and non-stable condition (Fig. 8), for the GR and EC methods
7. Stationarity criteria following the criteria of Foken and Wichura (Foken and Wichura 1996), for the for GR and EC methods
8. The intermittency was applied according to Mahrt et al. (1998), for the EC method
9. WPL correction due to water vapour and heat flux, for the GR and EC methods
10. Cross sensitivity of the compounds on the used analyzers, for the GR and EC methods
11. Preparation of the uncertainty budget for the measurement results, for the GR and EC methods

To that end it would also be good to quantitatively compare the two flux methods for CO_2, as it could help validating the gradient method. In this context I would expect to see a scatter plot and regression of both fluxes, - how well did the two methods really compare?

We agree with the comment. Unfortunately the fluxes of CO_2 were too small to be detected by the GR method. This can be seen from the concentration difference of the CO_2 between the measurement heights and shown in Fig. S2g. After the WPL correction applied to the measurement of CO_2 concentration the difference of CO2 did not exceed the calculated uncertainty limit. In the previous version of the manuscript this was not included into the results but at present it was corrected. We made the comparison of GR and EC methods with sensible heat, as shown in the figure below. The calculation of the sensible heat by GR method was conducted from the sea surface up the measurement height 11 m and to 15 m. The temperature difference between the measurement heights (11 and 15 m) were mostly too small to be detected.

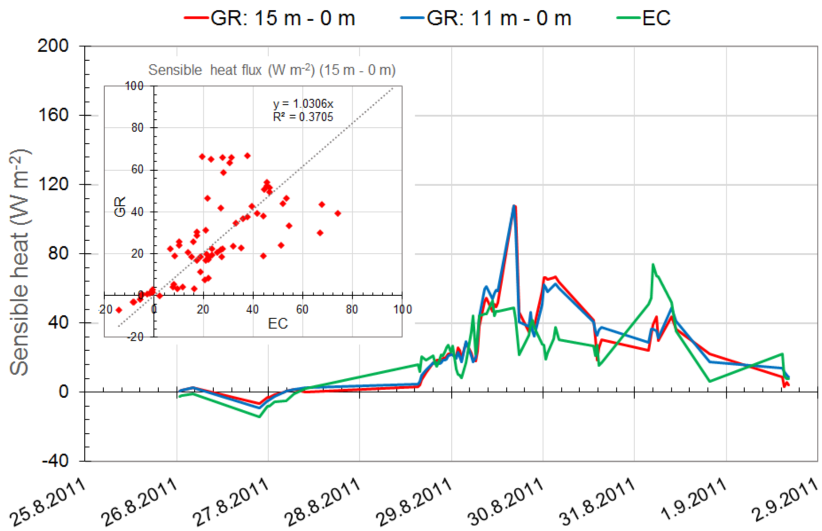

Fig.1. Sensible heat by GR and EC method at Harmaja 2011. The scatter figure between the two methods is shown in the small figure.

References

Foken, T, Wichura, B. Tools for quality assessment of surface-based flux measurements. Agric. Forest Meteorol. 78, 83 – 105, 1996.

Högström, U., Sahlée, E., Drennan, W. M, Kahma, K. K., Smedman, A.-S., Johansson, C., Pettersson, H., Rutgersson A., Tuomi, L., Zhang, F., Johansson, M.: Momentum fluxes and wind gradients in marine boundary layer – a multi-platform study, Boreal Env. Res., 13, 475-502, 2008.

Mahrt, L., Sun, J., Blumen, W., Delany, A.C, Oncley S. Nocturnal Boundary-Layer Regimes. Boundary-Layer Meteorology 88, 255 – 278, 1998.

Pirjola L., Pajunoja A., Walden J., Jalkanen J.-P., Rönkkö T., Kousa A., Koskentalo T.: Mobile measurements of ship emissions in two harbour areas in Finland, Atmos. Meas. Tech., 7, 149 – 161, 2014.