We are grateful to Reviewers #1 and #2 for taking time to comment on the manuscript. Thank you for the constructive criticism. The suggestions of the referees resulted in a clearer and better revised version of the original manuscript. Below, we provide a point by point response to the raised questions. For clarity, we first list *the referee comment,* and then our response (Authors' Response (AR)) in the following.

Referee #1: The only thing that concerns me is that the set-up is quite huge for only one compound, which can be, after couple of assumptions, measured by e.g. AMS. Why part of the sample was discarded to the waste when it can be directed to other analysis? Even if concentrations are too low, this "waste" could be collected to a longer loop or to a trap and analyzed with lower time resolution, for example by HPLC-MS. I understand that this can't be done anymore, so, authors can at least speculate about this issue.

AR: The referee is right that the sample solution can be directed to other analysis instruments as well, not only to waste. The authors have added a short discussion about the issue to the manuscript (Section 2.3.1): "In this study, most of the sample solution flow was directed to waste. However, it could be directed to other analysis apparatuses, such as ion chromatographs or a carbon analyzer, or it could be collected for offline analysis to gain more information about the sample."

Referee #1: 5500, 15-20: No need for such precise explanation

AR: The text was shortened based on the suggestion.

Referee #1: Section 2.3.1. Could sample loop be larger? How size was affecting the analysis?

AR: The authors have tested the different sample loop volumes in their previous study (Saarnio et al., 2010b). However, it was not stated in the published paper or here in this manuscript but the loop volume of 50 µL was optimized based on the sufficient amount of analytes for the detection from the diluted samples and at the same time without a remarkable effect on the broadening of the analyte peaks in the chromatographic separation. If the sample size was larger the peaks would broaden and, especially, in the case of concentrated samples broadening would cause overlapping of sequent peaks in the chromatogram. A sentence about the issue was added to the text: "The sample loop volume was tested to be suitable for separation of levoglucosan from two other MAs (Saarnio et al., 2010b)."

Referee #1: 5504, 1-5: Simultaneously? How? Explain, because it seems that the whole sample was continuously analyzed on-line. Or was it stopped during the collection of off-line samples?

AR: Part of the filter samples was analyzed during the maintenance breaks of the online system of PILS–HPAEC–MS and the rest of them were analyzed afterwards (after the measurement campaign). It was described in the original text that the filter samples were stored in freezer until the analysis (page 5503, lines 23-24). The text was changed accordingly: "The offline analysis was conducted with the HPAEC–MS using the same analysis program and apparatus as above presented for the online analyses. Part of the filter samples was analyzed during the maintenance breaks of the online system and the rest after the measurement campaign. The filter samples were stored in a freezer before the analysis."

Referee #1: 5506, 5-10: It's not a problem at all. Labelled compound is always a better choice.10-15: Levoglucosan is still added to PILS as standard addition, so, I don't understand statement in 5-10 about suppression.

AR: There were two reasons why we changed the internal standard from C13-labeled levoglucosan to methyl-β-D-arabinopyranoside: First, on the economic point of view the use of relatively expensive C13-labeled levoglucosan is not an optimum choice because the consumption of the internal standard is rather ample in the PILS–HPAEC–MS. Secondly, in our previous study (Saarnio et al., 2010b) where the HPAEC–MS method was developed, it was noted that simultaneously eluting compounds cause a slight ion suppression for both compounds. It means that the responses of C13-labeled levoglucosan in MS detection are changing depending on the amount of levoglucosan in the samples. That causes inaccuracy in the results. This issue was discussed in detail in Saarnio et al. (2010b). However, the main reason was money.

Referee #1: Section 3.2. 25-26 February seems odd. See fig. 4. You should discuss it as well.

AR: Actually, on 25–26 February there shouldn't be anything odd (online results 8% lower than offline). The variation of levoglucosan concentrations with the PILS–HPAEC–MS and from the filters are discussed now in more detail: "However, the ratio between the PILS–HPAEC–MS and filter results varied; e.g., on 24–25 February the levoglucosan concentration was 21% lower with the PILS–HPAEC–MS than from the filters, on 26–28 February only 1% lower and on 10–11 March 41% lower."

Referee #1: Fig. 8. About 68% is represented by one SD, two perhaps?

AR: In Fig. 8 vertical lines represent one SD, not two.

Referee #2: Page 5497 line 13: Anhydro monosaccharides should be anhydromonosaccharides, and the abbreviation should be 'AM'. Alternatively, if the authors prefer 'MA' as an abbreviation, it should be monosaccharide anhydrides.

AR: The text was modified as follows: "Anhydrosugars such as monosaccharide anhydrides (MAs: levoglucosan, mannosan, and galactosan, of which levoglucosan is typically the most abundant one) are generally used as biomass burning tracers in ambient air particles".

Referee #2: Page 5500 line 4: Sigma Aldrich should be Sigma-Aldrich. Is galactosan technical grade? Please provide the quality of galactosan.

AR: The name of chemical provider was modified as requested. Unfortunately the quality of galactosan was not told by the manufacturer and therefore it cannot be presented in the text.

Referee #2: Page 5500 line 12: Millipore is now Merck Millipore, and their corporate HQ is in MA, USA.

AR: The information about the company was changed according to referee's comment.

Referee #2: Page 5502 Determination with HPEAC-MS: I presume the authors used a negative ion mode. This information is missing in the description of the MS method.

AR: Yes, the referee is right. The information is missing in the text because the analytical method was originally referred to our previous publication (Saarnio et al., 2010b). However, we have now added the missing information in the text: "The ionization was made using electrospray technique (ESI) in the negative mode, and the mass range scanning (m/z 50–250) and selected ion monitoring (SIM) modes were used for the detection."

Referee #2: Page 5507 "Standard addition method": It is interesting that the authors added a levoglucosan standard solution to improve the detection and quantification of the ambient

samples. However, the authors state in page 5506 that the ISTD signals showed a significant variation when the method was tested for ambient aerosols, implying that the signals obtained from the standard addition likely showed a significant variation as well. Have the authors improved the variation by the standard addition? If not, have the authors determined statistically significant differences between the signals obtained from the standard addition and the ambient sample + standard addition? I.e. If the 100 ng/mL signal of levoglucosan standard addition has a standard deviation of 25% (say 100 25 ng/mL), what would be a criteria for the authors to quantify the ambient samples? 150 ng/mL levoglucosan signal? 200 ng/mL levoglucosan signal?

AR: The referee has pointed out an important topic that was not written clearly in the original text; the use of standard addition method adds an extra source of error. On the other hand, standard addition method enables the determination of low ambient concentrations.

The referee referred to page 5506 where it was mentioned that there was a lot of variation in the results of levoglucosan and ISTD. However, in the next sentence we noted that by increasing the concentration of ISTD the system showed better stability; i.e., there was less variation in the results of both levoglucosan and ISTD. What we want to say is that by increasing the concentration of ISTD, the variation of results could be decreased. The standard addition method was used primarily to expand the range of measurement – not to improve the variation.

Not any statistical analysis was made to find out the significance levels of variation of signals and concentrations. Therefore the authors have now removed the wording 'significant' from the parts of the text where one might confuse it with statistical analysis results. The following sentences were also added to the text (Sect. 3.1.3): "The standard deviation of the standard addition was 16%. The variation of the standard addition caused some additional inaccuracy in the quantification of ambient levoglucosan concentrations but it also enabled a wider range of measurement."

As the online determined concentrations of levoglucosan were compared with the biomass burning tracers analyzed with HR-ToF-AMS, it was noted that most of the high-concentration peaks were observed with both methods but there was a lot of variation in the small concentrations (Sect. 3.3). A mention about the uncertainty caused by the standard addition method was also added to this section: "There was a lot of variation, especially for

the small concentrations that was possibly caused by the inaccuracy of the standard addition method."

Referee #2: Page 5511 line 21: (1) The standard deviation values do not contain much information when they are larger than the average values. The ranges of levoglucosan concentrations should be given here instead. (2) Have the authors found statistically significant difference between the daytime and the nighttime levoglucosan concentrations? The difference in the average values does not necessarily mean that two sets of data are statistically significantly different.

AR: (1) The text was changed as recommended to contain information on the range instead of standard deviation. Also median values were added to the text: "In nighttime the levoglucosan concentration was on average only 8% higher than in daytime with the average levoglucosan concentrations (median; range) during the night and day being 86 (56; 0 – 1519) ng m^{-3} and 79 (57; 0 – 753) ng m^{-3}, respectively." (2) Not any statistical analysis was made for the data of daytime and nighttime levoglucosan concentrations. The authors are not stating that the found difference was statistically significant – quite the contrary, it was emphasized already in the original text that there was only a minor difference.

Authors' correction: In the schematic representation of the system (presented in Fig 1), the sampling inlet has been incorrectly named as PM_1 cyclone instead of PM_1 inlet. This will be corrected in the final version.