Final response to the referees and commenters

We thank all reviewers and contributors for their comments on our manuscript, in particular Dayle McDermitt who was able to constructively pinpoint a relevant issue that needs to be revised in our manuscript and which will strengthen and improve our paper. The reviewers mainly propose important modifications in terminological details that definitely will improve our manuscript but do not affect the content of our manuscript, including the conclusions, as we will explain our reply to the reviewers below.

The way how we referred to the WPL correction in the manuscript was rather unfortunate and left a completely wrong impression – our paper only referred to the WPL correction when it comes to the empirical approximation of the correction that is necessary to correct flux measurements effectuated with a closed-path methane analyzer of the type we used under absence of concurrent water vapor concentration measurements. To address this strong critique expressed by Reviewer #1 we will reorganize our material to make clear that we are not debating on the widely used WPL correction and hence will focus our work more on the spectroscopic effect that needs to be corrected for besides the generally accepted need for correction for density fluctuations. Namely, Section 2 which is now prominently headed as “Background” will be reduced and incorporated into “Material and Methods” to make clear that this is not the background of our work, but only a summary of what the generally used density flux correction suggests. To make clear that this density flux correction is not a perfect approach and that other arguments are around as presented by Reviewer #1 we will add a short paragraph with a critical discussion on the generally used approach to correct flux measurements for density fluctuations. Since this is not the key topic of our paper, we will make sure in our revisions that it becomes clear that this part does no longer appear to be the key message as Reviewer #1 wrongly assumed.

In the following, we assess the critical points raised by our reviewers and how we will incorporate their helpful suggestions into the revised manuscript. Where appropriate, we present the reviewer's comments in *italic* font. Our answers then follow in in normal font in the following indented paragraph.

Referee #1

The manuscript reflects a poor physical understanding. The authors try to suggest that the so-called Webb flux correction has somewhat to do with open-path and closed-path instruments. That is not the case.

> It is true, that the type of sensor does not necessary determine the type of the correction. Webb, Pearman, and Leuning (1980) stated under which measurement conditions which correction should be applied: “If the measurement involves sensing of the fluctuations or mean gradient of the constituent’s mixing ratio relative to the dry air component, then no correction is required; while with sensing of the constituent’s specific mass content relative to the total moist air, a correction arising from the water vapor flux only is required. Correspondingly, if in mean gradient measurements the constituent’s density is measured in air from different heights which has been pre-dried and brought to a common temperature, then again no correction is required; while if the original (moist) air itself is brought to a common temperature, then only a correction arising from the water vapor flux is required. If the constituent’s density fluctuations or mean gradients are measured directly in the air in situ, then corrections arising from both heat and water vapor fluxes are required.” (end of citation from WPL 1980). However, certain types of instruments imply that e.g. the sampled air cannot be brought to a constant temperature in case of an open-path instrument and hence these types of instruments necessarily require certain corrections. We will adjust

our wording to avoid the wrong impression that this reviewer had with our present text.

The papers of Webb & Pearman (1977) and Webb et al. (1980) do not reflect any specific application. These papers are only dealing with a correction regarding the air density. Following the arguments of Webb and co-authors, this correction should result in a mean vertical velocity, and it is suggested that this velocity contributes to an enhanced vertical flux component in case of trace gases like carbon dioxide.
From the beginning on, this flux correction was always a matter of an controversial discussion as reflected by many papers that can be found in the respective literature. The authors, however, completely ignored these discussion. Papers link that of Bakan (1978), Bernhard & Piazena (1988), Kramm et al. (1995), Kramm and Meixner (2000), Paw U et al. (2000), Fuehrer and Friehe (2002), Kramm and Dlugi (2006) are completely excluded. The review paper of Fuehrer & Friehe (2002) already discussed the pros and cons of this flux correction. Kramm & Meixner (2000) and especially Kramm & Dlugi (2006) argued that "the conventional Webb correction is based on elements of a Boussinesq approximation. Such elements, however, should not be considered while any kind of flux correction equation is derived because flux correction equations that are, completely or partly, Boussinesq approximated violate conservation laws like the equation of continuity and the balance equations for water vapor and trace species derived for turbulent systems." These are enough reasons for discarding the Webb flux correction. In front of this aspect, it is highly awkward to discuss the difference between open-path and closed-path instruments. The Webb flux correction is the problem, but not the kind of sensor.

We agree with the argument that the correction introduced by Webb et al. (1980) has been discussed and debated. Since the intention of our work was not to add to this debate, but rather to present an application of our findings, we started from the most frequently used version of the correction. The title as well as the “Background” section might have set our manuscript in a wrong light. As Reviewer #1 states, we do not question and test the WPL correction itself. Our aim was to show how the correction changes with the given implications as the WPL correction is widely used in the flux community. Other flux correction approaches could be modified similarly as we did in our manuscript. We respectfully acknowledge that other authors have presented other general recommendations on how to deal with flux corrections, as listed by Reviewer #1. Hence we will explicitly write that we suggest an empirical correction and not conduct a theoretical assessment. There are many examples available where a theory was considered still useful, although it has been shown to not be perfectly correct. The well-known philosopher Karl Popper names Newton's classical mechanics theory as one example, which still after its disproval by Einstein is considered a useful theory (although with a somewhat restricted domain of validity). Similarly, experimentalists have successfully used empirical corrections that are not perfectly agreeing with theoretical considerations – hence we will take care in our revisions to take this point of conflict out of the text since it received too much attention for what it actually represents in our manuscript.

In detail, in the revision of the manuscript, we will move the background section to the methods section and change the title (see answers to Referee #3) to detach the focus of the reader from the WPL correction itself. Additionally, we will add that the correction introduced by Webb et al. (1980) has been debated and other approaches exist. We also can offer to place a statement in our revised manuscript that says that improving the sensors would be the preferred future development over the improvement of correction functions that may be questionable due to their imperfect assumptions.

Referee #2 (Dayle McDermitt)

Specific comment #1:

We agree that the concept of dry mole fraction of water vapor is no physical entity for this special case. We also agree that the introduction of this unit would result in mathematical incorrectness as stated in Dayle McDermitt's review. Therefore, we carefully checked our calculations and verified that we actually did not calculate dry mole fraction of water vapor but used humid mole fractions, which means that we have the right entity but the wrong terminology, for which we apologize. This means that the presented results reflect the appropriate physical units, however were incorrectly named in the manuscript. We will correct the naming and also clarify the units of the measured properties in the method section. We thank Dayle McDermitt for addressing this important issue.

Specific comment #2:

It is true that the expression "observed flux in the cell" is unfortunate and might be misleading. We agree with the reviewer that the water vapor flux is not observed in the measurement cell of the instrument and that this basically reflects sloppy use of a precise terminology on our side. The measurements rather represent the water vapor fluctuations in the cell, which are consequently used to calculate the flux. We will reword the affected phrases to correctly reflect the circumstances.

Technical comments:

We will include all suggested technical corrections in our revised manuscript.

Referee #3

It is certainly interesting to assess whether it is possible to use an external H2O measurement for correcting spectroscopic closed-path CH4 measurements when H2O is not available from the spectrometer for eddy-covariance applications.

We thank this reviewer for this positive statement.

To do this, the (spectral) effects of sensor separation, dampening due to the tubing filters and instrument design, plus the specific tube delay effects for H2O and cross-sensitivity should
be determined. This does not require any modification of WPL theory, but only additional corrections and transfer functions. It is purely coincidental that two non-linear effects, i.e. cross-sensitivity and dampening effects, can compensate each other. Therefore, both should be treated separately and not merged into one correction.

Instead of applying the individual transfer functions, we determined the damping of the total signal using the cospectral correction model presented by Eugster and Senn (1995). Recently, Detto et al. (2011)* came to the same conclusion that one single correction function for all effects is an appropriate way to correct for damping losses (their equation (7)). This is an established and widely used procedure. We however agree that in our revisions we must be clearer in our wording to reflect the fact that these processes are not related at all with the WPL theory and can be considered by themselves. As stated in our conclusions, the spectroscopic correction and the damping of the water vapor signal did coincidentally compensate each other for our setup. In this aspect we agree with the reviewer and interpret his statement as a confirmation that he also thinks this might be coincidental – but this is an important statement for readers so they know the expected order of magnitude of this correction and could carefully compare whether their system is comparable to ours and could be corrected in the same way.

In the result section, we actually considered each process separately, but merged everything into one final empirical equation which might have raised the impression of a single correction. We will adapt the manuscript to stress that the final equation incorporates several corrections which only coincidentally cancelled out each other for our specific setup.

*Reference: Detto, M.; Verfaillie, J.; Anderson, F.; Xu, L. & Baldocchi, D. (2011) Comparing laser-based open- and closed-path gas analyzers to measure methane fluxes using the eddy covariance method. Agric. Forest Meteorol., 151, 1312-1324. doi:10.1016/j.agrformet.2011.05.014

If the goal of this ms is to find a way to make the FMA usable for eddy covariance this reviewer would disagree with the authors that the proposed approach is more elegant (p.353, l.22) than drying the sample.

We are open on the discussion on whether drying the sample before the measurement is the more elegant way than correcting for artifacts. The fact however is that drying the sample introduces new artifacts (see p. 353, l. 24–26 with the reference to Griffis et al. 2008 which also is confirmed by our own experience in previous studies). We hence will reword lines 26–27 on page 353 to avoid the dispute on elegance. We realize that our wording was not optimal and that we should rather neutrally state that in cases where drying is not possible or introduces adverse artifacts in the flux measurements, our proposed correction could contribute to solving the problem. Additionally, our approach can also be used to correct and recalculate older datasets where drying was not carried out.

If the goal is to make FGGAs without H2O correction (early serial numbers) usable for eddy covariance the relevance of this ms is not very general.

This is one aspect, but not the single goal of the manuscript. Nowadays, many groups use laser spectrometers to perform eddy flux measurements. In the latest FluxLetter (Vol.4 No. 3 January, 2012) more than 50 teams are listed who are performing methane eddy covariance measurements or are planning to start soon. Instruments of other manufacturers than Los Gatos Inc. also show spectroscopic effects. Since the instruments are commercially available, one might assume that the measurements are not affected or already corrected for spectroscopic effects. As it turns out, manufacturers are starting to get aware and improve their instruments, thanks also to scientific work as the one presented in our manuscript, which clearly helps to make producers of instruments aware of such artifacts.

But an even more important goal of this paper is to make researchers using such instruments aware of these effects. Even though our corrections are not very general, our suggestions can be used as a recipe for an analogue procedure and still have a high relevance for the target audience of our paper: researchers doing eddy covariance flux measurements of methane with this or similar instruments.

The authors failed to explain why the second H2O/CO2 instrument should be placed at some distance (p.354, l.7), in this case 5 m lateral and 0.95 m vertical. The vertical separation causes unnecessary problems for comparing the spectral response of both systems. The lateral separation limits the comparability in the time domain.

Two independent towers were used to perform the flux measurements and both consisted of a sonic anemometer and an as close as possible attached gas analyzer or inlet tube. We did not claim that the two towers “should” be placed at some distance, we neutrally described that one tower was about 5 m from the

other (p. 359, l. 15). We do not compute covariances with raw data time series from one sensor on one tower in combination with the ultrasonic anemometer of the other tower. We always use the pairs of instruments that are only separated by a short distance from each other (0.25 m below the sonic for the CH4 flux measurements as stated on p. 359, l. 2). We did not specify the distance of sensors for our conventional tower since we refer to Zeeman et al. (2010) for these details (p. 359, l. 16). Zeeman et al. specify "The EC sensor separation was 0.25 m, and the IRGA was tilted to the north to prevent incidence of direct solar light." We will add this information to our revised version since obviously simply referring to Zeeman et al. (2010) was insufficient.

If the reviewer wanted to express that computing covariances with sensors that are 5 m apart horizontally and 0.95 m vertically is not a good idea, then we can understand this feedback. We theoretically agree that the distance of the two towers would be critical for the comparison of high frequency data, but since we did not do this nor even attempt to do this, we consider the reviewers concern as an important misunderstanding that we need to avoid with our revised text.

We only use the H_2O flux measured on one tower to approximate a density flux correction for the other tower. We consider this a sound approach since the fetch at the measurement site was large enough such that the different footprints due to the different instrument heights did cover the same homogeneous ecosystem surface. We successfully carried out flux comparisons using 30-minute averages with two towers which were separated by a few meters in earlier studies (see Eugster et al. 1997*).

*Reference: Eugster, W.; McFadden, J. P. & Chapin, III, F. S. (1997) A comparative approach to regional variation in surface fluxes using mobile eddy correlation towers. Boundary-Layer Meteorol., 85, 293-307.

The character of this experiment is rather a case study and that should be represented in the title, the abstract and throughout the text of the ms. A more suitable title could be "Can open-path H2O measurements be used for correcting closed-path spectrometer without internal H2O measurements?" Parts of the conclusion are in line with this already (p. 371, l. 8-10).

We agree that the title as well as the background section raised wrong expectations and do not represent the real content of our study. We are happy to change the title according to the reviewer's suggestion.

Although the topic and the points raised in the discussion about instrument interference and usability of instruments for eddy-covariance applications are insightful and valuable, I recommend rejecting this ms.

We are not happy with this suggestion, but think that we are able to revise our manuscript to adequately address the scientific critique expressed by this reviewer. At the same time we respect that this reviewer has some opinions that differ from ours.

Contributor #1 (Patrick Sturm)

Your paper nicely shows that a careful characterization of commercial laser spectrometers is still indispensable for accurate and reliable field measurements and I very much enjoyed reading it.
Allow me to point out a detail with respect to the damping loss correction which from my point of view is slightly confusing. On page 361, lines 13-15 you write: "A slight modification of the theoretical cospectral model introduced by Kaimal et al. (1972) is

damped according to the inductance L [s^{-1}] in an alternating-current circuit to match the calculated cospectra." My concern here (apart from the incorrect syntax and the wrong unit) is the term "inductance". The analogy with an electric circuit is a very useful concept to describe the damping of the cospectra. Eugster and Senn (1995), which you are referring to in the paper, showed that the damping can be nicely described in analogy with a low-pass RL-filter. However, the characteristic of such a first-order filter is described by a time constant and in case of the RL-filter the time constant is given by τ = L/R. Thus, L as it is used here actually corresponds to the time constant L/R [s] and not to the inductance (which has units of Vs/A). When describing the damping of a first-order system (be it an eddy covariance instrument or an electric RL-filter) I would therefore suggest to use the term "time constant" and the symbol τ instead of L.

We agree that the term inductance is confusing as even in the analogy of an alternating electric circuit, the first order damping corresponds to the time constant τ=R/L and not the inductance L. However, the term “time constant” might be confusing as well since the damping depends on the atmospheric conditions, mainly the relative humidity (see Fig. 2 in the discussion version of the manuscript). Hence, we rather suggest introducing the term “time factor” for the symbol τ and change the unit to [s], as we already did in a previous paper by Eugster & Zeeman (2006)*. We will also adapt the method section correspondingly and check the units and symbols in the whole manuscript.

*Reference: Eugster, W. & Zeeman, M. J. (2006) Micrometeorological techniques to measure ecosystem-scale greenhouse gas fluxes for model validation and improvement. International Congress Series, 1293, 66-75.

Two other details I stumbled upon:
Page 356, line 10: "relatet" should be "related".

Will be changed.

Page 358, line 7: I guess you have assessed the accuracy rather than the precision by using the NOAA standards.

Indeed, accuracy was meant. We will clarify this in the revision of the manuscript.