Answers to the comments of anonymous Referee number # 1 to Gerilowski et al., "MAMAP – a new spectrometer system for column-averaged methane and carbon dioxide observations from aircraft: instrument description and performance assessment.", Atmos. Meas. Tech. Discuss., 3, 3199–3276, 2010

First of all we would like to thank the referee for the detailed, helpful and competent comments. Below we present our response and clarifications to each of these comments. As the reviewer itself provided no numbering for his comments, we introduced a numbering of the major and minor referee comments and hope that it reflects the referees view.

## **Major comments:**

#M1: The manuscript is very long and it would clearly benefit from shortening the text and removing unnecessary details. There are many sections that appear very repetitive. In particular the introduction section with 9 pages appears to be unnecessary long and it discusses several publications from other authors in great detail. At the same time, several figures and the details shown in the figures are too small (Fig 5 to 11)

R#M1: We agree with the comment of anonymous referee #1. The introduction section will be substantially shortened for the revised version of the manuscript and the plots will be enlarged as possible.

#M2 The manuscript really only deals with an assessment of the precision for the CO2/CH4 ratio and the authors argue that this is the most relevant quantity for estimating sources of hot spots. Of course it is true that using the CO2/CH4 ratio will significantly reduce most biases, but it can still be expected that the assumption of the retrieval, instrument calibration or atmospheric variations can cause significant biases that can vary on scales of kilometers e.g. due to the coupling with surface albedo. When attempting to estimate emission strength, it is of critical importance that the uncertainty of the estimate is well characterized and traceable. The last section of section 6 goes into this direction, but is very brief and incomplete. I would suggest that the authors include a critical discussion of the different error sources and the expected effect on the final product.

R#M2: As the main focus of the paper is to introduce the instrument and its performance, a detailed error budget w.r.t. emission strength estimation is out of scope of the paper. As mentioned in section 5 a detailed discussion on the impact of aerosols, thin cirrus clouds. albedo etc. on the accuracy of MAMAP data products will be given in Krings et al. (manuscript in preparation). Errors of the instrument calibration like uncertainty of the slit function, wavelength calibration and retrieval assumptions as for example a-priori aerosol profile used for the RTM, will primarily lead to constant biases on the retrieved column. The main remaining parameter on small scales (i.e. some kilometers) is variation of albedo with aerosol (and thin cirrus) coupling. For this study background aerosol variations and variations of cirrus clouds are assumed to be small during the target overflights under stable (clear sky) atmospheric conditions what in many cases (i.e. landfills, dry seeps, mud volcanoes, venting etc) is a reasonable assumption when aerosols are not produced in large amounts by the source itself. The main varying parameter in that case is the (measured) detector signal variation which is primarily connected to variations of the albedo. Equation (25) gives an upper error estimate for signal dependent variations of the retrieved CH4/CO2 column concentration and therefore includes beside effects like albedo dependent aerosol and cirrus coupling also instrumental effects like for instance detector nonlinearity. When large amount of aerosols are produced by the source (i.e. power plants, steelworks, volcanoes etc) a detailed in case error analysis needs to be performed. In Krings et al. (manuscript in preparation) estimates/inversion models including in depth error analysis of the power plant CO2 emissions based on MAMAP data will be presented. We will add a short paragraph listing other instrumental error sources affecting the estimate of emissions at the end of chapter 6. It has also to be pointed out, that fast varying albedo variations on small scales are accounted for in the precision estimate as such effects can not be separated from other fast varying effects as mentioned in section 4

#M3: In addition, it needs to be pointed out clearly that the described approach will either give CO2 or(!) CH4 columns, but not both. The instrument has the potential to deliver both, but this would require a very different retrieval approach and the presented performance assessment would not be valid in this case.

R#M3: We agree, that the approach presented in this paper delivers XCH4 or XCO2 i.e. column averaged dry air mixing ratios (mole fractions). The statement will be included in the revised version for this approach. However it has to be mentioned that most local anthropogenic sources like power plants, landfills, fugitive emissions from refineries and gas processing plants, steelworks etc. produce primarily either CO2 or CH4. It can be shown by the equations given in section 5 that for instance the emission estimate of a medium size CO2 emitting power plant will barely be affected by CH4 emissions of the surrounding wetlands. With the same equations it can be shown, that the amount of CO2 emitted by a landfill which has the same or a factor of ~ 2-3 bigger magnitude as the amount of the emitted Methane will not affect the methane emission estimate because of the more than 200 times higher background concentration of CO2 (a small paragraph with this additional calculations will be added in section 6). As mentioned by the anonymous referee #1 the instrument is designed and has the potential to deliver both XCO2 and XCH4 namely by the use of the simultaneous measured O2A column. This approach will require a similar SNR and Precision analysis as performed in this paper but for the CO2/O2 and CH4/O2 ratios. In this paper we focus primarily on the performance analysis of the SWIR cannel. A performance analysis of the O2A band SNR and precision and the according XCO2(O2) and XCH4(O2) SNR and precision will be presented in a separate publication.

#M4: Furthermore, as mentioned in the text, the instrument would be very interesting for satellite validation, but this would require that this observations from this instrument itself are validated.

R#M4: We generally agree with the referee #1 in this point and validation of the instrument is planned to be performed in the near future (i.e. for instance comparison to ground based FTIR measurements etc.). In addition, combined remote sensing and in-situ flights are planned where emissions estimates of "Hot-Spots" performed with the in-situ payload will be compared with emission estimates performed by MAMAP.

## **Minor Comments:**

#m1: Title: '...performance assessment' It is not clear to me what is meant by performance assessment. The manuscript really only deals with SNR and precision.

R#m1: We agree. The word "assessment" will be removed/exchanged in the title.

#m2: Abstract: ... in-situ local point measurements from micro sites... ->What do you mean by micro-sites. Are there in-situ macro-sites ?

R#m2: We agree, this will be changed from "in-situ local point measurements from micro sites" to "microsites" only as microsites imply always in-situ. Microsites are sites where emissions are estimated on micro scales (i.e.  $0.1-100~m^2$ ) for instance by closed chamber measurements (see Sachs et al., Environmental controls on CH4 emission from polygonal tundra on the microsite scale in the Lena river delta, Siberia, Global Change Biology (2010), doi: 10.1111/j.1365-2486.2010.02232.x). Using this definition we believe that eddy co-variance towers measuring on local and ecosystem scale ( $10^4-10^6~m^2$ ) and in-situ tall tower sites measuring on ecosystem and mesoscale can be defined as "in-situ regional sites" or "in-situ macrosites".

#m3: ...for these scenes the CH4 and CO2 column retrieval precisions are typically about 1% -> This manuscript only deals with CO2/Ch4 ratios and not with simultaneous CH4 and CO2 column observations. You should make this point very clear.

R#m3: We agree. This will be changed to CH4 / CO2 column retrieval precision ratio and the point will be clarified (see also #M3)

#m4: The measurements by the MAMAP sensor enable estimates of anthropogenic, biogenic and geological emissions of localized intense CH4 and CO2 sources ->... couldenable...

R#m4: We agree. This will be changed.

#m5: Introduction: This arises in part because of the difficulty in estimating the highly variable natural and anthropogenic atmospheric source emissions in space and time -> This arises in part because of the difficulty in estimating the highly spatially and temporally variable natural and anthropogenic atmospheric source emissions.

R#m5: We agree. This will be changed.

#m6: Up to now, flux estimates of CH4 and CO2 in current global... -> Up to now, flux estimates of CH4 and CO2 based on current global...

R#m6: We agree. This will be changed.

#m7: ...on precise atmospheric in-situ... -> on precise and accurate atmospheric in-situ

R#m7: We agree. This will be changed.

#m8: ...of the surface observation site network... ->...of the surface observation network...

R#m8: We agree. This will be changed.

#m9: The footprint of the Tanso-FTS instrument is 10 km... -> The footprint of the Tanso-FTS instrument has a diameter of 10 km...

R#m9: We agree. This will be changed.

#m10:...as they combine range with high spatial resolution. ->... as they combine coverage with high spatial resolution.

R#m10: We agree. This will be changed.

#m11: Until recently, few airborne instruments have had the capability of measuring CH4 atmospheric columns with the high spatial resolutions... -> Until recently, few airborne instruments have had the capability of measuring CH4 atmospheric columns with high spatial resolutions...

R#m11: We agree. This will be changed.

#m12:....reach a threshold sensitivity during flight at typical flight... -> ...reach a threshold sensitivity during flights at typical flight...

R#m12: We agree. This will be changed.

#m13: In order to improve our understanding of CH4 and CO2 sources (and sinks) and their variability at micro and meso scales by using top-down constraints and to validate space-based measurements on meso and synoptic scales (i.e. from SCIAMACHY and GOSAT), new measurement systems are needed. These must be capable of measuring CH4 from high altitudes (>7 km) over different surface types at high horizontal 15 resolution (<250m) over areas <10 km up to 200 km and yield a precision and accuracy equal or better than the accuracy achieved by current and planned sensors, i.e. with a precision and accuracy of equal or better than about 1–2% (Bréon and Ciais, 2010) -> I don't think that the cited publication talks about requirements for airborne systems and I am wondering how there requirement have been inferred. A requirement on precision and accuracy of 1-2% is very large and I would argue that a regional bias of 2% will make it impossible to learn anything on CO2 fluxes.

R#m13: We agree that a 2% relative accuracy is insufficient to learn anything on natural CO2 fluxes. As the instrument was initially designed to measure methane, the threshold and goal requirement is inferred primarily for methane emitters, citation: "The measured data ideally needs to be of an accuracy and precision to yield on inversion the CH4 emissions from less intense but extensive and larger scale sources and sinks, such as wetlands. As a threshold the accuracy and precision of the data yields on inversion significant constrains on local hot spot emissions to separate them from the less intense but extensive larger scale sources and sinks and thereby allowing an improved estimate of

both." i.e. goal <= 1 %, threshold = 2%. Nevertheless the threshold criteria is inferred primarily for methane it would allow also flux estimates on strong anthropogenic (and potentially also of geologic CO2 emitters). Thus the instrument can potentially be used for independent monitoring. For validation of current satellite sensors the (validated) instrument (relative) accuracy and precision needs to be equal or better than that of current available satellite sensors. This text section will be modified for the revised version.

## #m14: Description of the MAMAP instrument

I am surprised that not more details are provided here. How is the ILS measured and how does it change across the detector. Has there been any attempt to measure absolute calibration? Do you measure the non-linearity of the system? Is there stray light in the system?

R#m14: The ILS of the detector was measured by a SLS. As the SLS provides only a single line in the SWIR the motorized grating was slightly rotated in different positions to estimate the line shape over the entire detector. The measured asymmetric ILS shape then can be sufficiently good fitted by 2 Gaussian functions. The FWHM of the fitted ILS was further optimized by slightly adjusting the FWHM while minimizing the fit residua. Nonlinearity measurements were performed and provided by the camera manufacturer (we use commercial spectroscopy cameras for MAMAP). We checked the linearity values provided by the manufacturer by some linearity tests in our lab using a stable WLS and a narrow thermally stabilized band pass filter in combination with changes of the exposure time of the instrument. A real linearity test with the required accuracy for fixed exposure times and adjusted light levels will require a sophisticated optical set-up currently not available in our lab (required setups are available at the national metrology organizations i.e. NIST (USA) or PTB (Germany)). Stray light measurements and absolute radiometric calibration measurements have actually not been performed. Absolute radiometric calibration is not necessarily required for the currently used retrieval algorithm as albedo is actually not derived from the data. It is plant to perform a full instrument characterization and radiometric calibration during this winter. Nevertheless the spectrometer throughput (without lenses and folding-mirrors) was measured in the lab with a calibrated Spectralon -NIST FEL lamp setup. Instrument calibration parameters were inferred (with limited accuracy) from these measurements and knowledge of the lens throughput and the mirror reflectivity (derived from manufacturer data). The inferred parameters compared to ZEMAX model simulations of the instrument. We will add a few sentences to the manuscript according to the issue.

### #m15: The O2-A-band NIR spectrometer

Since this manuscript only deals with CO2/CH4 ratios, the details of the O2 A Band detector and how it will be used together with the SWIR instrument are somewhat less relevant. Nevertheless, how is co-boresighting of both spectrometers achieved and how well can it be characterised.

R#m15: Co-alignment of both spectrometers in along-slit direction is achieved by imaging a small slit in infinity and aligning both slits with respect to that slit and also by moving s small source inside the field of view of the spectrometers. As the ground scene size along track is primarily defined by the co-adding and exposure time (see section 2.2), small misalignments of both slits in cross slit direction are not critical. The co-alignment in along slit direction is achieved digitally (see section 2.2) by moving the co-adding window of the

O2-A band while minimizing the variations of the retrieved O2A column with respect to the retrieved CH4 and CO2 columns. Focusing of both spectrometers was performed by using a solar simulation unit described in Gurlit et al., The UV-A and visible solar irradiance spectrum: inter-comparison of absolutely calibrated, spectrally medium resolution solar irradiance spectra from balloon- and satellite-borne measurements, Atmos. Chem. Phys., 5, 1879–1890, 2005.

#m16: MAMAP retrieval algorithm and the determination of data products

"There are several approaches to estimate the dry air column needed for the conversion of the greenhouse gas columns into column-averaged mixing ratios: ->Method (II) is exclusively used here and this should be pointed out clearly and thus only CO2/CH4 ratios are retrieved and CO2 and CH4 cannot be determined simultaneously"

R#m16: Method (II) is "using another well mixed gas whose mixing ratio is well enough known and varies less than the trace gas of interest" and is not completely new. It has been used to derive XCH4 mole fractions by means of the simultaneously retrieved CO2 column. Using it vice versa in case of CO2 hot-spots has also been proposed in Bovensmann et al, 2010, and seems to be highly appropriate e.g. in case of hot-spots. Simultaneous retrieval of CO2 and CH4 by MAMAP is possible e.g. by using also the O2A channel information as has been done e.g. for SCIAMACHY satellite instrument and will also be done for using CO2 information from OCO. However this is not within the scope of this publication, since for hot-spots the CH4/CO2 or CO2/CH4 ratios have the potential to be more accurate. Reasons are named and quantified in Bovensmann et al., 2010. See also authors response to referee #1 comment #M3.

#m17: "An alternative approach for the determination of XCH4 at least in regions where diurnal or spatial CO2 variations are small is to assume that the CO2 is effectively constant and well mixed compared to CH4 -> Usually it is not assumed to be constant but a modelled CO2 is used"

R#m17: For the most CH4 hot-spot applications (landfills etc.) variations of the CO2 total column over and around the source with significant impact on the measurements are not to be expected as can be easily shown by the equations given in section 5. However it is true that this has to be carefully checked in detail for every single application before the ratio is applied and evaluated. Modeled CO2 is used for SCIAMACHY but actually not used for MAMAP as on scales of 0.1° and below there is currently a lack of model data.

#m18 "the path of the electromagnetic radiation is approximately identical -> the path of the electromagnetic radiation is similar..."

R#m18: We agree. This will be changed.

#m19: "In summary all three methods can be used for MAMAP. The method, which performs best, depends on the target and the validity of the assumptions and the effort made to account for cloud and aerosol within the retrieval algorithm. -> ...all three methods could..."

R#m19: We agree. This will be changed.

#m20: Method (III) is not really applicable as it would require knowledge of surface pressure on a scale of a few meters"

R#m20: We agree. This will be changed.

#m21: "Method (I) would require at least very good co-boresighting of both spectrometers and some level of absolute calibration to obtain estimates of surface albedo."

R#m21: We agree. This will be changed. (radiometric calibration is currently not required as no albedo correction has been applied in the current version of the algorithm, see also authors response to referee #1 comment #m14)

#m22: "obtained in the SWIR (1.6 nm) -> ...obtained in the SWIR (1.6 micron)"

R#m22: We agree. This will be changed.

#m23: "...with respect to a change of atmospheric parameters (mainly scaling factors for the CH4, CFO2, and H2O vertical profiles and temperature profile shift) -> please provide a complete set of all retrieved parameters"

#### R#m23:

Retrieval of CO2:

geophysical parameters: - CO2, - H2O, - Temperature

nongeophysical fit parameters: - low order polynomial accounting for surface spectral reflectance, radiometric calibration function, etc., - odd even correction fitting an alternating function [-1, 1, -1, 1, ...] (odd even effect caused by the multiplexer design of the SWIR detector in combination with tilted illumination)

## Retrieval of CH4:

geophysical parameters:- CH4, - CO2, - H2O, - Temperature

nongeophysical fit parameters: - low order polynomial accounting for surface spectral reflectance, radiometric calibration function, etc., - odd even correction fitting an alternating function [-1, 1, -1, 1, ...]

m#24: "In addition to the geophysical fit parameters a low order polynomial in the spectral domain is used to account for all smoothly varying spectral parameters, which are not explicitly modelled or inadequately known. These parameters include, for example, the MAMAP absolute radiometric calibration function, aerosol scattering, and absorption parameters and the surface spectral reflectance -> It needs to be pointed out here that these assumptions can be made because only CO2/CH4 ratios are considered here and it is assumed that biases introduced by these assumptions will ratio out. But even in this DOAS approach, you need to make some assumptions about surface albedo and aerosol loading for the radiative transfer calculations."

R#m24: For the reference scenario used for the retrieval we assumed a constant surface albedo of 0.18 and a standard LOWTRAN background aerosol scenario as used in Schneising et al, 2008. This will be mentioned in the revised version of the publication. In future version of the algorithm also an albedo correction based on radiometric instrument calibration will be applied.

#m25: "This is currently attributed to wavelength calibration ... -> How is the wavelength calibration carried out? Do you fit shift and stretch?"

R#m25: Wavelength calibration has been performed by the absorber features itself using shift and squeeze of the fit procedure. Because of the spectral oversampling of 8-9 pixel/FWHM this approach delivers sufficiently good results for MAMAP. Once the initial wavelength calibration parameter are retrieved only shifting was applied during data retrieval to account for shifts caused by inhomogeneous illumination. We also performed tests by fitting the spectral shift in combination with the FWHM of the line shape but these did not improve the data quality. Switching the shift parameter off (no shift, no squeeze) leads to a slightly decrease of SNR and precision in comparison to retrievals where shift fitting was applied.

#m26: "Inspection of the averaging kernels (Fig. 6)... -> Please define averaging kernels"

R#m26: A definition of averaging kernels will be given in the revised version of the paper.

#m27: "SCIATRAN accounts for this path doubling in the weighting functions for each layer -> I would assume that you cannot simply double the path, but you need to use 1+1/cos(SZA) as a factor"

R#m27: "Doubling" was an unfortunate choice of words. Of course the RTM SCIATRAN takes into account SZA and measurement geometry. The revised paper will be more clear regarding this point.

#m28: "Hence a column averaged PSF will always overestimate the real concentrations in the total column, because the averaged weighting functions are smaller than the weighting functions below the aircraft. -> I have real difficulties to comprehend why such an approach has been chosen, e.g. the new BESD retrieval would clearly be much better suited for the analysis of the data."

R#m28: BESD is a new algorithm currently under development for retrieval of SCIAMACHY satellite data and has not yet been applied and tested to MAMAP. For the assessment of the SNR and precision of the instrument we preferred the more simpler WFMD/M algorithm as the behavior of the algorithm with respect to different combinations of free fit parameters was extensively tested for MAMAP in the last years. Implementation of BESD would also require absolute radiometric calibration of the instrument which is currently not performed.

#m29: "The correction factor c will surely depend on more than altitude, SZA and albedo. What about the VMR profile shapes, the aerosol loading, the water vapor column,

temperature profile or potential cloud coverage above the aircraft altitude? Where is the surface albedo taken from? Finally what is the estimated uncertainty of the parameter c?"

R#m29: The correction factor was assumed to be constant for this study (for a given aircraft altitude and SZA) and is here basically the inverse of the mean averaging kernel below the aircraft. In depth accuracy analysis of the MAMAP measurements is not within the scope of this study, which focuses on the instrument and not so much on external parameters which include the mentioned aerosol, surface spectral reflectance, cloud coverage etc. A detailed discussion of the uncertainties of the conversion factors will come along with the in depth analysis of the power plant measurements in Krings et al., which is in preparation. Please note, that a 1% error on the c-factor alone would only result in a 1% error in the retrieved column variation (not the total column) and thus in a 1% error of the emission rate, which is well below current uncertainties given in literature for any CO2 or CH4 sources.

Please note further that an error has been found in the compilation of Table 2. Values for the c-factors of an aircraft altitude of 1250m and 4500m are affected. The corrected table can be found below. The new correction factors slightly improves our CH4 precision estimates for 4500 m, SZA 40°, by a factor of 0.869 (albedo 0.01) and slightly downgrade the values for same altitude and SZA but for albedo of 0.18 by a factor of 1.0585. The identified error does not affect the general results and conclusions of our analysis.

Affected are in detail total column accuracy values calculated on page 3231, line 3 & 5, page 3232, line 12 & 15, page 3234, line 20 & 25 & 26 page 3245, line 11 & 14, page 3245, line 25 & 26, page 3246, line 21

Airplane	Solar Zenith	Surface	Conversion Factor [-]	
Altitude [m]	Angle [°]	Albedo [-]	CH₄	CO <sub>2</sub>
850	40	0.01	0.575	0.459
850	40	0.05	0.546	0.435
850	40	0.18	0.535	0.426
850	50	0.01	0.607	0.478
850	50	0.05	0.571	0.448
850	50	0.18	0.558	0.438
1250	40	0.18	0.556	0.447
4500	40	0.01	0.729	0.619
4500	40	0.05	0.712	0.604
4500	40	0.18	0.706	0.599
4500	50	0.01	0.747	0.626
4500	50	0.05	0.727	0.609
4500	50	0.18	0.720	0.603

Corrected Table 2.

#m30: Performance assessment of the SWIR channel In the following sections the terms precision, accuracy and measurement uncertainty are used. ->accuracy is not considered in this manuscript

R#m30: The term "accuracy variation & relative accuracy" is used several times in the following sections of the manuscript. There is a need for a definition what is mean by the

term accuracy in this manuscript. In the revised version also a definition of the term accuracy variation or relative accuracy as used in the manuscript will be added.

#m31: Theoretical retrieval precisions Eq (13): The Gaussian error propagation for a ratio a/b will yield: sqrt( var(a)/b2 + var(b)\* a2/b4 )

R#m31: As both columns for the theoretical analysis are normalized to background concentration (i.e. a=1 and b=1) Eq (13) directly follows from sqrt(  $var(a)/b2 + var(b)^*$  a2/b4). This will be clarified in the revised version.

#m32: To account for small systematic offsets caused by the fit procedure... ->What kind of systematic offsets do you mean?

R#m32: Systematic biases caused for instance by insufficient knowledge of the slit function used for the RTM simulation. This small systematic biases are (partly) accounted for by this normalization (see also authors response to referee #1 comment #M2).

#m33: Ground based measurements: In a second on-ground set-up, scattered light zenith radiance measurements (measurements of the down-welling diffuse radiance) were performed... ->How is this data analyzed without knowing the aerosol load?

R#m33: We assumed a standard LOWTRAN background scenario for the aerosol load for the RTM simulation. We believe that this approach delivers sufficient results for the precision and SNR estimate in ground based Zenith viewing geometry.

#m34: Airborne single readout measurements over inhomogeneous scenes Effects such as small spectral features of the earthshine spectral reflectance can also not be completely excluded as reason. ->Which kind of effect is meant here? The same statement is repeated in the section 4.3.4.

R#m34: As the earth's surface is a type of surface diffuser, spectral feature effects like described for surface diffusers laboratory measurements (see. Bazalgette et al. Proceedings of the Second Working Meeting on MERIS and AATSR Calibration and Geophysical, Enhancement of Diffusers BRDF accuracy, Validation (MAVT-2006), 20-24 March 2006, ESRIN, Frascati, Italy (ESA SP-615, July 2006) ) can not completely be excluded as limiting factor. We believe that such effects will be minimized by the introduction of the spatial scrambler unit. Additionally, at the best of our knowledge, high resolution spectral libraries for different materials have not yet been published for the investigated spectral region. Thus interferences of the DOAS retrieval with material spectral features also can not completely be excluded.

#m35: For a background concentration 1774 ppb this corresponds to an enhancement oft 20.59 ppb of the total column. ->For a background concentration 1774 ppb this corresponds to an enhancement of 20.59 ppb of the total column.

R#m35: We agree. This, will be changed.

#m36: Below 1% total column precision, the accuracy variation induced by atmospheric effects (i.e. light path differences for CH4 and CO2 caused by scattering and absorption of aerosols and clouds and variations of the albedo/SSR and refractive index of the atmosphere) start to dominate the overall uncertainty variation. ->This statement implies that the relative accuracy is better than 1%. I would assume that the c-factor alone is uncertain by more than 1%. Please add evidence for this statement since accuracy is not addressed in the manuscript.

R#m36: Please note, that the c-factor is applied to the total column increase and not to the retrieved total column. Thus an error of the c-factor of 1% would result in a 1% error of the total column increase and not in a 1% error of the retrieved total column (see also authors response to referee #1 comments #M2 and #m29)

## #m37: MAMAP targets

Achieving 1–2% total column accuracy.... Nevertheless, such a performance puts limitations on the target emissions which are suitable to be detected with the actual MAMAP total column precision of 1%. ->Again, it is assumed that the accuracy of MAMP is much smaller than the precision without providing any evidence.

R#m37: The term "accuracy" will be changed to "relative accuracy" in this context. It will be stated, that this is a reasonable assumption for the case of cloud free stable atmospheric conditions and changes of the albedo within a given interval as can be derived from equation 25. It will be additionally stated, that the accuracy of MAMAP will be discussed in detail in Krings. et. al. (see also authors response to referee #1 comment #M2)

#m38: In addition assuming that aerosol and cloud are smoothly varying, any impact of slow systematic changes in the accuracy of the measured CH4/CO2 column mixing ratio can be minimised by high-pass filtering the data. ->I would argue that many biases will be on a similar scale than the plumes that are targeted by MAMAP, in particular effects of aerosols and clouds will vary with surface albedo which will vary on a range of different scales. Please show that data with and without this filter.

R#m38: For the generation of the plume plots presented in this paper a high-pass filter with a 80 point (80 measurements) window was used applied to the co-added burst mode measurements ( $PSFR_{BM}$ ). Thus only low frequency variations were removed from the dataset of the whole ~ 2 hour flight. The high-pass filter window was ~ 2.6 - 3 times bigger than the measurements taken for single plume transects (i.e. < ~ 25-30 measurements). The whole flight was processed in one shot with one RTM for fixed altitude and fixed SZA. All 3 presented 2D-plots are clip-outs of this single processed data set. A high-pass filter can be applied to the data but is not necessarily required if only data-points from overflights around the source are processed separately. It will be added to the description of the plots of Fig 12,13 and 14 that the presented data is currently filtered with a 80 point high-pass filter primarily removing low frequency variations.

#m39: Over the globe, many different sources exist with fluxes exceeding the above calculated detection limits. -> It is not really about the detection of emissions, but about measuring their emissions with more accuracy than is done already. Thus, the considerations on flux estimates from MAMAP need to have some realistic uncertainties.

R#m39: This paper does not really deal with inversion of fluxes. Inversion of fluxes with error analysis will be presented in Krings et. al.. Nevertheless also the simple approach presented in this paper (section 5 and 6) can be applied to constrain fluxes of Methane in several circumstances as Methane emissions from localized sources can often have uncertainties of more than 40-100 % (i.e. landfills, gas industry, etc). As mentioned in the paper, Chambers et al. reported for instance a factor of up to 4 - 9 difference for bottom-up and top-down emission estimates of single gas and oil industry facilities. (see also authors response to referee #1 comment #M2)

#m40: The approach used above can also be applied to estimate the detection limits required for extended regions of less intense source emissions assuming that the region is sufficiently homogeneous. ->The assumptions of the presented methods in this manuscript will be poor for larger regions and I would expect that for larger regions, small biases will become the dominant error source.

R#m40: This was clearly stated at the end of the section: "This simple method to estimate the range of expected CH4 total column changes emanating from these type of sources requires sufficiently stable conditions during the period of the aircraft measurement. When this requirement is not fulfilled, more complex regional chemical transport modeling (Jagovkina et al., 2000, 2001) is needed. For extended sources the smallest detectable flux Fmin is restricted primarily by accuracy variations (i.e. the relative accuracy) and not by the precision of the instrument. A detailed discussion on the impact of aerosols and thin clouds on the accuracy of MAMAP data products is given in Krings et al. (2010)." (see also authors response to referee #1 comment #M2)

#m41: First results from measurements over localized emissions sources In order to test the MAMAP sensitivity to score emissions and validate the results obtained in Sect. 4.3.4 flights over localized targets have been performed ->I don't think that the result shown in this manuscript qualifies as validation.

R#m41: We agree. Tthe word "validation" will be exchanged by the word "test" or "check"

#m42: Systematic effects in the measured columns caused by solar zenith angle changes and cirrus cloud variations are minimised by high-pass filtering the data ->Why does the solar zenith angle introduce biases. As already mentioned above, I don't think that high-pass filtering is appropriate and it would be beneficial to show the unfiltered result as well.

R#m42: Actually the whole flight is processed with a single RTM simulation for one altitude and a fixed solar zenith angle. Thus a change in SZA of several degrees during the flight will produce a changing bias as can be shown by RTM simulations. (see also authors response to referee #1 comment #m37)

#m43: Figure12/13: Albeit the high-pass filter, the Co2/Ch4 ratio still shows significant variations outside of the expected plume. Do you attribute this to atmospheric variations, instrumental issues or retrieval algorithm issues? Can this be used as a measure of the accuracy?

R#m43: This question can be answered first after optical modification of the instrument to account for inhomogeneous slit illumination. If then a precision smaller than the relative accuracy can be achieved small scale relative accuracy variations can be better separated from the precision errors. Currently the observed variations can be attributed to both atmospheric signals by the surrounded industry complex or to precision errors currently limited primarily by the illumination conditions of the slit (see SNR and precision discussion in section 4 & section 7). Small scale relative accuracy errors are currently included in the precision estimate as they actually can not be separated from precision effects (see also definition of precision: "As systematic fast varying (near random) albedo/SSR effects can not be separated from the other fast random effects, they also are accounted for in the precision.")

#m44: ... in agreement with the predicted values -> What is the predicted value?

R#m44: We agree. Predicted values will be added.

#m45: To exclude albedo/SSR dependent offsets as origin of the CH4 column increase.... ->How good is the fit and the correlation coefficient? ->It would be interesting to do the same test for the other flights shown in figures 12 and 13.

R#m45: The fit is calculated from the high-pass filtered data of the whole flight including data shown in Plots 11, 12, and 13. The correlation coefficient will be added ( Cc~ 0.2 ). This correlation coefficient is not very high but gives a first guess for the signal (and those also for the albedo/SSR) dependency of the retrieved CH4/CO2 data also including other signal depended effects like for instance non linearity of the detector.

#m46: -> Does this explain all the variations in outside of the wind direction from the source?

R#m46: See authors response to referee #1 comment #m43.

# #m47: Summary

In that case the MAMAP CH4 total column uncertainty variation will no longer be limited by the precision of the instrument but by the relative accuracy ->Again, there is no evidence in the manuscript that this is the case.

R#m47: The complete citation in the manuscript was :

"After modification of the optical bench, total column precisions << 1% for ground scene lengths (LT) <200m are predicted. In that case the MAMAP CH4 total column uncertainty variation will no longer be limited by......". Thus this statement implies that the accuracy variation is larger than "<< 1 %" what is a reasonable assumption. See also referee # 1 comment #m35 where the referee assumes that the expected relative accuracy can be bigger 1 % which is similar to the statement of the authors above.

#m48: MAMAP measurements can potentially be used for micro-, meso- and synoptic scale validation of daily CH4 and CO2 chemical transport model simulations, and for validation of satellite measurements... ->this manuscript only deals with CO2/Ch4 ratios

and it will be a completely different story to obtain accurate, independent and simultaneous CO2 and CH4 column data.

R#m48: See authors response to referee #1 comment #M2 & #M3.