Foreword:

We would like to thank the reviewers for their comments, which have helped improving the paper. We have addressed all of the issues that they have raised. Changes are documented hereafter and in the text. Reviewer comments are shown in black and answers are highlighted in blue. A brief summary of the relevant changes made to the manuscript is presented after answers to anonymous referee #2 and is followed by a version of the manuscript that highlights in blue the changes that have been made.

Anonymous Referee #1

The paper describes a high-resolution version of the AirCore, a device designed to take samples throughout parts of the stratosphere and the full troposphere, and analyse them for CO2 and CH4 by retaining much of the information contained in the vertical profile structure. The paper is mostly well-written, and fits well within the scope of AMT. However, a number of issues listed below should be addressed before the paper can be recommended for publication.

General comments:

[1] Regarding the design criteria for the AirCore-HR, a bit more details should be given. Does diffusion/dispersion during sample analysis play a role?

Dispersion and diffusion during sample analysis play a role, which is taken into account. Diffusion alone is considered during the waiting time before analysis and both diffusion and dispersion are considered during analysis time.

The description has been changed to include in details the roles played by diffusion and dispersion. (See page 4, line 27 – page 6, line 10)

[2] Does the drying cartridge cause diffusion, what is the volume of the cartridge? How does this affect the design of the AirCore, i.e. the lengths of the tubes?

The role of the drying cartridge as contributing to diffusion is not considered. It is a very small volume (10 cm ¼ inch, partly occupied by magnesium perchlorate). The additional volume is very small and represents less than 0.005% of the total volume.

This has been added section 2.2 (page 7, line 25-26 as follows: "The additional volume to the system is very small and represents less than 0.005 % of the total volume (it is thus not considered in the calculation the vertical resolution (sect. 2.1.1))"

[3] Uncertainty analysis is well done in general, in that the contributions from different sources of uncertainty to the overall uncertainty of the profiles of CO2 and CH4 are assessed separately. However, the potential effects on CO2 from the chemical dryer of the AirCore-HR should be quantitatively estimated, and if possible an upper limit should be given for the loss of CO2. If there are on the order of 20 ppm CO2 missing due to a 7-hour interaction of the sample with the magnesium perchlorate contained in the dryer, the quality of the rest of the CO2 profile is

rendered questionable. What is the estimated loss of CO2 during normal flow conditions? What is the volume of the dryer, what amount of magnesium perchlorate was used? Also the difference in CO2 from AirCore-HR and AirCore-GUF, which is up to 3 ppm and far beyond the uncertainty estimate for AirCore-HR, should be properly assessed in this context. If the chemical dryer has such an impact, the data should not be published.

The aim of the uncertainty analysis presented here is to assess the uncertainty related to the processing and measurement of the sample. The potential effect of the dryer on CO_2 mixing ratios is a separate issue that remains to be solved. As described in the paper, the agreement between different AirCores in terms of vertical structures is good, but a bias still remains. However, for CH_4, no issue remains. We think that this difference between CO_2 and CH_4 should be known by the community, and this is why we have chosen to chosen to keep all the results.

Moreover, differences in CO_2 are being investigated to understand whether the effect comes from the drying agent or from other possible effect with the tubing. For future campaigns in 2017, we have designed specific tests to assess the impact of the dryer such as flying several AirCores without dryers.

Additional text mentioning the difficulties with CO_2 and way forward has been added in the abstract (Page 1), in the conclusion (page 18) and in section 4.1 (while describing the profiles) (page 12 – lines 18-26).

For the volume of the dryer, see answer above.

Specific comments

[4] Pg 1 Line 12: may be reformulate "The multi-instruments gondola also carried two other . . ."

 Sentence has been modified according to recommendation. (now Pg 1 Line 12)

[5] Pg 2 Line 33: I suggest adding "for deployment on high altitude balloons as platform" after "have been developed"

Sentence has been modified according to recommendation. (now Pg 3 Line 3-4)

[6] Pg 4 Line 1: In Karion et al. (2010) there is no mentioning of avoiding turbulent flow as being a criteria, it just turned out to be laminar given other constraints on flow and tube diameter.

Yes. Given the AirCore characteristics the flow is always laminar. To answer a similar question raised by reviewer n°2 a paragraph has been added. (see Pg 4, Line 10-27)

[7] Pg 4 Line 12: A reason for not considering diffusion and dispersion during the air sampling process (i.e. the ascend of the balloon) should be given.

During the ascent the tube empties and the initial fill gas that has the same concentration in the whole volume of the AirCore exits. It is not yet the sampled air that is inside the tube and thus there is no reason to consider this phase. Sampling starts in the descent phase.

During the sampling process that occurs in the descent phase diffusion is not considered because ambient pressure constantly evolves. The repartition of the sampled ambient air varies all along the descent and it is only once at the surface that the sampled volume is fully compressed into the tube. This is when the diffusion along a given distance will translate into the highest impact on vertical resolution. We make the assumption that the role of diffusion and dispersion during the sampling phase is minor compared to the waiting phase before analysis and the analysis phase itself.

The paragraph has been modified in the paper to detail this (See page 4, line 28 – page 6, line 10).

[8] Pg 4 Line 15: This factor two is not included in Karion et al. (2010), it should be clearly introduced and justified

It is a matter of notation; the factor two comes from considering diffusion in both directions. Karion et al. (2010) decided to include it in the final step when calculating the vertical resolution. Anyway, to avoid confusion, the same notation as in Karion et al. (2010) is now used and the additional factor 2 appears in equation 8 (Pg 6)

[9] Pg 4 Line 16: Eq. 3 is a completely different equation, not just a different expression of Eq. 2.

The phrasing was incorrect. The sentence has been modified (see Pg 8 Ln 1-3):

« Using equation 7 and knowing that air is distributed in the AirCore as a linear function of total pressure column sampled, it is possible to evaluate the pressure range affected by mixing related to diffusion and dispersion. »

[10] Pg 5 Line 17: the curve for the original AirCore shows 1 km (not 2 km) resolution at 20 km altitude

Yes. It has been corrected and additionally Figure 2 and 3 were modified as we improved the calculation of the vertical resolution using a standard atmosphere from the TIGR database to calculate the pressure/altitude relationship rather then an isothermal atmosphere. See (Pg 6, Line 7-10)

Sentence is now Pg 6 Line 30: "After 3 hours of waiting time before analysis, it is possible to achieve a vertical resolution of 250 m at 10 km and 1,2 km at 20 km."

[11] Pg 5 Line 226: I suggest rewording "to flow under" with maybe "be carried by"

Sentence has been modified according to recommendation. (now Pg 7 Line 3).

[12] Table 1: It would be useful to also include the corresponding numbers for the standard AirCore and the AirCore-GUF. Data for inner radius seem redundant, as the diameter is given. I assume with "width" the wall thickness of the tubing is meant.

Redundant information about the inner radius has been deleted but the data about the other AirCores hasn't been added as the paper focuses on the presentation of AirCore-HR.

[13] Pg 6 Line 18: What is the meaning of "only" in this context? Does it point out that no humidified standard air has been used?

It just meant that the calibrated standards that allow correcting to the WMO scale contained dry air. Sentence has been modified by deleting the "only". (now Pg 8 Line 4).

[14] Pg 7 Line 19: It is not the drift due to water vapour, but the combined effect from dilution and line broadening caused by water vapour that the correction compensates for in the reported dry air mole fractions.

Second reviewer made a similar comment, also recommending to cite Chen et al. 2010 as well as Rella et al. 2013. The sentence has been modified as follows:

"These are automatically corrected by the instrument for a combined effect of dilution and line broadening caused by water vapor (Chen et al., 2010; Rella et al., 2013). Then, in a first processing step, the measured mixing ratios are corrected for a bias from the Picarro measurement to the WMO scales " (see Pg 9, Lines 6-8)

[15] Pg 7 Line 29: the sentence starting "Using the ideal gas law:" is not really a sentence.

Sentence has been modified as follows: "Using the ideal 15 gas law (equation 9), it is possible to calculate the number of moles captured in the tube all along the trajectory." (See page 9 Lines 19-20)

[16] Pg 8 Line 24: may be replace "It took part to" with "It participated in"Pg 8 Line 25: "consisted in" -> "consist of"

Sentence has been modified according to recommendation. (now Pg 10 Ln 14-15)

[17] Pg 10 Line 25: "exchange rate . . . takes several years" this need reformulation. Also an earlier reference for deriving mean age Boering et al., 1996: Boering, K. A., Wofsy, S. C., Daube, B. C. and Schneider, H. R.: Stratospheric mean ages and transport rates from observations of carbon dioxide and nitrous oxide, Science, 1996.

Earlier reference has been added (Page 12 Line 12)

[18] Figure 8: rather than not showing the data between 70 and 90 hPa I suggest showing them in a different color.

Figure was modified to present the complete profile. The parts of the profiles affected by unrealistic values are presented in dotted line. See Figure 12.

[19] Pg 11 Line 22: I suggest not adding 24 ppb, as rather than "easing" the comparison it will confuse the reader. Why not doing an honest comparison? The range of the x-axis in Fig. 9 (b) (90 ppb in the inset, and 1200 ppb for the main figure) should allow for enough room to show both profiles without alteration.

The purpose here is to focus on the structures themselves. Since the removal of 24 ppb is clearly

stated in the text, we find this comparison honest. However, to avoid any confusion, the removal of the bias has been specified also in the legend of the figure. See figure 9 and text (page 13, Line 11-13) has been changed as follows:

"24 ppb were added to the CAMS-ECMWF CH_4 high resolution forecast to emphasize the good agreement on structures, rather than focusing on the bias which may be attributed to incorrect surface fluxes or issues with airmass exchanges along the vertical."

[20] Pg 11 Line 33: there is some discussion on this in a paper that has just appeared in ACPD: Verma, S., Marshall, J., Parrington, M., Agustí-Panareda, A., Massart, S., Chipperfield, M. P., Wilson, C. and Gerbig, C.: Extending methane profiles from aircraft into the stratosphere for satellite total column validation: A comparative analysis of different data sources, Atmos. Chem. Phys. Discuss., 1–32, doi:10.5194/acp-2016- 704, 2016.

Thank you for pointing this out, a citation to the paper was added. (Pg 13 Line 24)

[21] Pg 11 Line 9: It should be explained why the three temperatures measured are ex-pected to represent the uncertainty. How have they been measured exactly, and where along the tube; is it expected that the three sensors give different reasons e.g. due to an expected temperature gradient? Or is the assumption that the uncertainty will not be larger than the range, and the uncertainty is conservatively estimated?

The uncertainty is not larger than the range so this is the conservative way of including temperatures in the uncertainty estimates.

Text had been changed as follows: "Indeed, the 3 temperature probes are placed at different positions along the tube (near the entrance, in the middle of the AirCore and near the closed end) and depending on the distance to the inlet they have recorded different temperatures along the AirCore. Choosing randomly between one of the 3 probes is thus the conservative way accounting for the uncertainty related to the mean temperature of the AirCore." (Pg 13 Line 35 – Pg 14 Line 3)

Moreover, even with this conservative estimation of the temperature uncertainty, the impact of temperature variations on the overall uncertainty is very small. A paragraph has been added in Section 4.2 (page 14 line 30 – page 15 line 2)

[22] Pg 13 Line 3: please reformulate, e.g. use "single measurement point"

The sentence has been modified according to suggestion (now Pg 14 Ln 30)

[23] P14 Line 1, Figure 12 a): As before, I also suggest here showing the CO2 data from the AirCore-GUF analysis above 200 hPa, but in a different color.

Figure was modified to present the complete profile. The parts of the profiles affected by unrealistic values are presented in dotted line. See Figure 12.

Text was modified accordingly: "Therefore, all the CO_2 sampled above 100 hPa with AirCore-GUF was probably altered by the combination of unrealistic low values of CO_2 acquired during

the plateau phase and the diffusion effects. This part of the profile that should not be considered is shown in dotted line as with AirCore-HR CO2. Only AirCore-GUF CO2 sampled in the troposphere below 100 hPa should be compared, where residual effects from this phase are minimal." (Page 15 Line 28-32)

[24] P14 Line 7: Between 300 and 800 hPa the AirCore-GUF CO2 values are constantly above those of the AirCore-HR, mostly by more than one ppm, i.e. by significantly more than the claimed uncertainty. This cannot be explained by "only at a different resolution". Can the authors exclude that the AirCore-HR CO2 data are more impacted by a loss of CO2 in the dryer?

AirCore-HR and AirCore-GUF dryers don't have the same volume. AirCore-GUF values may seem higher between 300 and 800 hPa but in the new figure 12 you can notice that AirCore-GUF also shows unrealistic low values of CO2 above 200 hPa and that CO2 values are then lower than those from AirCore-HR. It is thus not as simple as incriminating one or the AirCore. Again, the impact of dryer needs to be further studied.

Anonymous Referee #2

Membrive et al. present AirCore-HR that is a version of AirCore with higher resolution than the originally one developed in Karion et al., and further provide comparisons of vertical resolutions of different AirCores. AirCore-HR was flown in the StratoScience 2014 campaign along with a lower resolution AirCore, which enables the authors to compare the resulting profiles in terms of vertical resolution and absolute values. Be- sides these, the authors have also estimated the uncertainties of the vertical profiles, and compared the observed profiles with model simulations.

The manuscript focuses on the design and comparison of vertical resolution of Air- Core profiles, which is a very useful aspect for improving the AirCore technique. The manuscript is well structured and written. I suggest publication after addressing the following points.

General:

[1] There is a lack of understanding of the disagreement in absolute values of CO2 from AirCoreHR and AirCore-GUF. The speculation on the dryer given by the authors is not convincing. See the detailed comments below.

Answers are provided below. Specific texts have been added in the paper to address this comment.

[2] How has the cell volume affected the vertical resolution? It was mentioned in section 2.1.1. However, it is not clear how this was taken into account in the calculations.

The description of the impact of diffusion, dispersion and volume of the cell on the vertical resolution was modified to present the various contributions in more details. See pages 4-5

Detailed comments:

[3] Page 4/line 6: how is the flow determined as being laminar? Can the authors give the upper limit of the flow rate for being laminar in the AirCore-HR?

A new paragraph has been added to detail the calculation of the laminar flow using Reynolds number. (See page 4 Lines 8-28) :

"The transition between laminar and turbulent flow can be evaluated thanks to the dimensionless Reynolds number (Reynolds, 1883). Taking into account the useful parameters to describe the tubing of an AirCore Reynolds number can be expressed as follows:
…

In all circumstances the flow in an AirCore is thus laminar because R_e is much inferior to 1750."

[4] Page 7/line 2: analyzers → analyzer Page 7/line 19: version → mole fraction

Sentences have been modified according to recommendations (see Page 8 line 20 and Page 9 line

7)

[5] Page 7/line 19-20: it is not for "an eventual drift of the measurement", but for dilution and spectroscopic interferences, see Chen et al., 2010 and Rella et al., 2013.

This was also pointed out by the first reviewer and has been modified. It is now described as follows :

"The dry mole fraction of CO_2 and CH_4 provided by the Picarro are used. These are automatically corrected by the instrument for a combined effect of dilution and line broadening caused by water vapor (Chen et al., 2010; Rella et al., 2013). Then, in a first processing step, the measured mixing ratios are corrected for a bias from the Picarro measurement to the WMO 5 scales." (Page 9 / lines 7-9)

[6] Page 10/line 31 – Page 11/line 3: These are too speculative. Magnesium perchlorate is widely used in ground-based and aircraft measurements of greenhouse gases. Levin et al. 2002 Tellus page 699 explored the potential effects of magnesium perchlorate on CO_2 measurements, and from the two experiments no significant biases due to the use of magnesium perchlorate were found.

This section has been reformulated to include other hypothesis, now page 12 / lines 18 to 26 : "The origin of these very low values of CO_2 is still debated. The alteration of CO_2 sampled in this range was possibly caused by the dryer. Indeed, the magnesium perchlorate used as drying agent at the entry point of the tube is slightly reactive with CO_2 and inert with CH_4. Because of the long exposure (~7h) to this drying agent during the plateau phase, enhanced by air coming in and out of the tube because of oscillations of the gondola around 80 hPa, the magnesium perchlorate might have impacted CO_2 sampled by the AirCore-HR. The dryer being inert to CH_4, no impact is seen on CH_4 profile. Other explanations might be that the air sampled during this particular phase was polluted through interaction with polystyrene, with the balloon envelope (pumping up some of the ambient air as they re-equilibrate with ambient air during this long phase) or by chemical interaction with helium from the balloon (during some short re-ascent phases)."

[7] Page 12/line 7: why not use accuracy instead of using precision? Note that the precision can be averaged down to even lower values when vertical resolution is considered.

Sentence has been modified according to recommendation (see Page 13 line 31)

[8] Page 14/line 11: stonger → stronger?

Sentence has been modified according to recommendation (see Page 16/ Line 4)

[9] Page 15/line 10-14: If it were explained as remaining impacts of diffusion with the very low values of altered CO_2 sampled at the plateau, AirCore-GUF would be expected lower than AirCore-HR; however, the observed difference is opposite. Furthermore, diffusion will unlikely have such an impact on the tropospheric values. I do not get the point why humidity plays an

increasing role. Please explain.

To answer a suggestion by reviewer n°1 the complete profiles are now shown on figures 8 and 12 and dotted lines represent the part of the profiles with unrealistically low values that should not be considered.

Now that the full profiles are available, it is clearly visible that bias between AirCore-GUF and AirCore-HR in terms of CO_2 is negative from the top of the profile till almost 200 hPa and then it becomes positive in the lower troposphere. It is because of the variable bias, once positive and once negative that we suspect it could be related to the dryer depending on the individual characteristics of each one (flow encountered, efficiency to remove humidity).

Anyhow the paragraph was changed to:

"The reasons of these observed difference are still debated. The main hypotheses are that it could be related to some kind of ''memory effect' of the tubing to the previously stored calibrated gas or it is also suspected that the individual dryers from different AirCores may affect the CO_2 samples slightly differently when capturing CO_2. Overall, the problem highlights that there are some remaining questions with CO_2 sampling and that some potential interferences with CO_2 have to be studied more closely." (page 17 / Lines 10-14).

[10] Table 2: How are the uncertainties defined? They seem to be unrealistically small.

Uncertainties associated to the mixing ratios of the target gases come from a set of measurement compared to other target gases analysed for WMO scales at LSCE. It is the standard deviation over 3 measurements measured with 5 days interval. It shows the reproducibility of the measurements (3 measurements over a 15-day period. It was performed at LSCE with a Picarro G2401 calibrated with a scale of 6 tanks from NOAA/ESRL.

Some text has been added to the paper to describe this see (table 2). Page 28

Relevant changes made in the manuscript

- The description of the impact of diffusion and dispersion on the vertical resolution has been completely revised (section 2.1.1) (Pages 4-6). It now includes a section describing how to calculate that the flow is laminar in the AirCore as well as fully detailed description of the equations that allow taking diffusion, dispersion and additional mixing in the volume of the cell of the analyzer into account to calculate the expected vertical resolution.
- Previous figures 2 and 3 showing the vertical resolution have been changed in order to take into account an improved version of the program to calculate the vertical resolution using an average temperature profile corresponding to temperate air masses typically found at mid-latitudes rather then an isothermal temperature profile.
- Previous figures 8 and 12 have been modified to include full CO2 profiles. The altered part of the CO2 data is now presented in dotted lines.
- A paragraph describing the effect of temperature uncertainty on the overall uncertainties has been added (Page 14 line 30 – Page 15 Line 2)
- Additional hypotheses have been added: (i) to explain the loss of CO_2 during the long plateau (Page 12 Lines 18- 26), (ii) a variable bias in other parts of the profile (Page 17, Lines 10-14).
- Issues remaining with the CO_2 profiles have been detailed in the abstract, in the manuscript and in the conclusion.

AirCore-HR: A high resolution column sampling to enhance the vertical description of CH_4 and CO_2

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Abstract. An original and innovative sampling system called AirCore was presented by NOAA in 2010 (Karion et al., 2010). It consists of a long (> 100m) and narrow (< 1cm) stainless steel tube that can retain a profile of atmospheric air. The captured air sample has then to be analyzed with a gas analyzer for trace mole fraction. In this study, we introduce a new AirCore aiming at improved resolution along the vertical with the objectives to: (i) better capture the vertical distribution of CO_2 and CH_4,

(ii) provide a tool to compare AirCores and validate the estimated vertical resolution achieved by AirCores. This AirCore-HR (high resolution) consists of a 300 m tube, combining 200 m of 1/8 in.(3.175 mm) tube and a 100 m of 1/4 in. (6.35 mm) tube. This new configuration allows to achieve a vertical resolution of 300 m up to 15 km and better than 500 m up to 22 km (if analysis of the retained sample is performed within 3 hours). The AirCore-HR was flown for the first time during the annual StratoScience campaign from CNES in August 2014 from Timmins (Ontario, Canada). High-resolution vertical profiles of CO_2 and CH_4 up to 25 km were successfully retrieved. These profiles revealed well defined transport structures in the troposphere (also seen in CAMS-ECMWF high resolution forecasts of CO_2 and CH_4 profiles) and captured the decrease of CO_2 and CH_4 in the stratosphere. The multi-instruments gondola also carried two other low-resolution AirCore-GUF that allowed to perform direct comparisons and study the underlying processing method used to convert the sample of air to greenhouse gases vertical profiles. In particular, degrading the AirCore-HR derived profiles to the low resolution of AirCore-GUF yields an excellent match between both sets of CH_4 profiles, and shows a good consistency in terms of vertical structures. This fully validates the theoretical vertical resolution achievable by AirCores. Concerning CO_2 although a good agreement is found in terms of vertical structure, the comparison between the various AirCores yields a large and variable bias (up to almost 3 ppm in some parts of the profiles). The reasons of this bias, possibly related to the drying agent used to dry the air, are still being investigated. Finally, the uncertainties associated with the measurements are assessed, yielding an average uncertainty below 3 ppb for CH_4 and 0.25 ppm for CO_2 with the major source of uncertainty coming from the potential loss of air sample on the ground and the choice of the starting and ending point of the collected air sample inside the tube. In an ideal case where the sample would be

fully retained, it would be possible to know precisely the pressure at which air was sampled last and thus to improve the overall uncertainty to about 0.1 ppm for CO_2 and 2 ppb for CH_4.

1 Introduction

Understanding the global atmospheric budget of the two major greenhouse gases (GHG) emitted by human activities, carbon dioxide (CO_2) and methane (CH_4), is essential for predicting their future concentration levels. To that end, several efforts have been dedicated to improving the monitoring capabilities of these gases. Under coordination from the World Meteorological Organization (WMO), a global atmospheric CO_2 and CH_4 monitoring network of surface-based stations has been established (GCOS, 2011) to provide continuous information on their atmospheric concentrations. Although essential to infer surface fluxes, these surface measurements are sparse and lack information pertaining to the vertical structure of the atmospheric CO_2 and CH_4. In order to improve spatial coverage, several satellite-based missions have been developed to monitor greenhouse gases from space. Observation in the short-wave infrared (SWIR) enable retrieving total atmospheric columns, during daytime, and mostly over land. SWIR missions include the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) spanning 2003–2012 (Frankenberg et al., 2011; Wecht et al., 2014), the Greenhouse Gases Observing Satellite (GOSAT) since 2009 (Hamazaki et al., 2007; Butz et al., 2011) and more recently the Orbiting Carbon Observatory (OCO-2) for CO_2 only (Crisp et al., 2004; Hammerling et al., 2012). Observations of terrestrial radiation in the thermal infrared (TIR) provide information mostly on mid-tropospheric columns, by day and night, over land and sea. Missions include the Atmospheric Infrared Sounder (AIRS) since 2002 (Crevoisier et al., 2003; Xiong et al., 2010), the Tropospheric Emission Spectrometer (TES) from 2004–2011 (Worden et al., 2012) and the Infrared Atmospheric Sounding Interferometer (IASI) since 2007 (Crevoisier et al., 2009a, b, 2013; Xiong et al., 2013). Vertical profiles of CO_2 and CH_4 are also available from limb measurements such as from the Atmospheric Chemistry Experiment (ACE-FTS) (Foucher et al., 2011). These satellite based vertical profiles mainly cover the upper troposphere and low stratosphere with a low vertical resolution.

One of the main challenges for any satellite-based measurements is data evaluation and the comparability to WMO standards. To that end the Total Column Observing Network (TCCON) (Wunch et al., 2010) has been established. It consists in a network of upward looking Fourier Transform Spectrometers (FTS) and has been widely used to evaluate retrievals of total columns from SWIR space missions (e.g. Houweling et al., 2014). TCCON provides column-averaged retrievals that do not have any vertical resolution and also require independent evaluation of the data.

Precise and regular vertical profile measurements from the surface to above the tropopause are currently missing to evaluate total or partial columns of GHG retrieved either from the ground or from space and to tie them to the calibrated measurements of the WMO.

Several aircraft missions contribute to provide vertical information with regular measurements along commercial airlines such as the CONTRAIL project (Machida et al., 2008) and the CARIBIC project (Schuck et al., 2009). Other less regular aircraft campaigns are also dedicated to study GHG at a local scale (Zhang et al., 2014; Chen et al., 2010; Karion et al., 2013;

Crevoisier et al., 2006, 2010; Sweeney et al., 2015) or from pole to pole such as the HIPPO project (Wofsy, 2011). Such vertical profiles are usually limited to 12 km.

To overcome this limitation, several instruments to measure CO_2 and CH_4 profiles have been developed for deployment on high altitude balloons. Commonly used techniques include Fourier Transform spectrometer (FTS) measurements such as the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) (Oelhaf et al., 1991), cryogenic samplers (e.g Schmidt and Khedim, 1991; Engel et al., 2008) to capture air in flasks at different altitudes along the balloon flight to be analyzed at a later stage, and laser diode spectrometers such as the Spectromètre Infra Rouge pour l'Etude de l'Atmosphère par Diode Laser Embarquées (SPIRALE) (Moreau et al., 2005) or Pico-SDLA instruments (Durry et al., 2004; Ghysels et al., 2011; Joly et al., 2007). All these instruments require to be flown on heavy balloon-borne platforms. They can thus not be flown on regular basis.

In this context, an original and innovative atmospheric sampling system called AirCore has been developed at the National Oceanic and Atmospheric Administration Earth System Research Laboratory (NOAA/ESRL) (Karion et al., 2010) from an idea originally developed and patented by Pieter Tans (Tans, 2009). It consists of a long and thin stainless steel tube shaped in the form of a coil which can sample the surrounding atmosphere and preserve a profile. This new system allows balloon measurements of GHG vertical profiles from the surface up to approximately 30 km. The vertical resolution is ultimately determined by the length and diameter of the tubes.

Since the development of the first AirCore (Karion et al., 2010), new and lighter AirCores have been developed at NOAA, Groningen University and Goethe University Frankfurt. These lighter AirCores capture a smaller volume of air leading to a slight decrease in the achievable vertical resolution. This paper focuses on the development of an AirCore that allows retrieving profiles of GHG with a higher resolution along the vertical, with the following objectives: (i) to better capture the vertical distribution of atmospheric CO_2 and CH_4, in the troposphere, UTLS and stratospheric regions, (ii) to provide a tool to compare low resolution AirCores and validate the theoretical resolution achievable by AirCores.

The design of this new high-resolution AirCore, named AirCore-HR, is presented in section 2 together with the description of experimental settings and processing method. Section 3 describes the first flight of AirCore-HR. Section 4 describes the resulting CO_2 and CH_4 high resolution profiles, associated uncertainties, and compares high and low resolutions profiles retrieved from 2 AirCores. Section 5 gives the conclusion.

2 AirCore-HR design, experimental setup and processing method

The general principle of an AirCore is illustrated in Fig.1. First, in a preparation phase, the tube is filled with a calibrated gas standard. It is placed under a balloon with one end of the tube open and the other end closed. During the ascent phase, as the air in the tube equilibrates with ambient pressure, the initial fill gas evacuates. After reaching an upper limit, where only a small fraction of the initial fill gas remains in the tube, the AirCore starts a descent phase. During this phase, as it maintains pressure equilibrium along the descent, the tube samples the ambient air. On the ground, the tube is closed, retaining the sampled profile to be analyzed with an analyzer for trace gas mole fraction.

2.1 Relation between AirCore design and vertical resolution

AirCores can be designed in a variety of configurations that determine the vertical resolution that can be achieved with the instrument. The resolution directly depends on the molecular diffusion and shear flow diffusivity otherwise known as Taylor dispersion inside the tube (Karion et al., 2010). The two major criteria in designing an AirCore are thus: (i) keeping the tubes diameter sufficiently thin to have a laminar flow at the sampling flow rates; (ii) constraining the total weight to fit the specific flight requirements of their carrier (weather balloons, stratospheric balloons, planes...) while allowing for sampling of a sufficient amount of air for the planned analysis.

2.1.1 Impact of diffusion and dispersion on the vertical resolution

As described in Karion et al. (2010), at the flow rates of gas entering the AirCore during flight (<235 sccm (standard cubic centimeters per minute)) and during analysis (30 to 120 sccm) the flow in the AirCore-HR is expected to be laminar. The transition between laminar and turbulent flow can be evaluated thanks to the dimensionless Reynolds number (Reynolds, 1883). Taking into account the useful parameters to describe the tubing of an AirCore Reynolds number can be expressed as follows:

$$R_e = \frac{Q d_{in}}{\nu \Sigma_{in}} \tag{1}$$

Where Q is the flow rate expressed in $m^3.s^{-1}$, d_{in} the inner diameter in meters, ν cinematic viscosity ($\frac{\mu}{\rho}$) in $m^2.s^{-1}$ and Σ_{in} the surface of the inner disc of a section of the tube m^2.

Most of the AirCore configurations will have the following characteristics: (i) ν the cinematic viscosity of air : 15.6 x 10^{-6} at 20 °C, (ii) the inner diameter of the tube d_{in} = 0.15 to 1 cm, (iii) Σ_{in} the surface of an inner disc of a section of the tube $2\pi r_{int}^2$ with r_{in} = 0.075 to 0.5 cm, (iv) the flow rate Q $\approx$ 0.7 $cm^3.s^{-1}$ during analysis (about 40 $cm^3.min^{-1}$), and possibly up ti 250 $cm^3.min^{-1}$ which is equivalent to $\approx$ 4 $cm^3.s^{-1}$ during the fast descent phases.

Such values yield a number of Reynolds between :

$$1,36 < R_e < 8,5 \tag{2}$$

In all circumstances the flow in an AirCore is thus laminar since R_e is much inferior to 1750 (Peixinho and Mullin, 2006).

Diffusion and dispersion are not considered during ascent while the tube empties, nor during descent where the sampled pressure range varies continuously and repartition of the air along the AirCore thus evolves rapidly. It is only from the moment the total column is sampled and the final air repartition reached that the described model is used to calculate the vertical resolution (i.e. from the moment the tube is sealed with the captured sample until the end of the analysis).

At first, during a given storage time before the payload is recovered only molecular diffusion will affect the sample. As described in Karion et al. (2010) the root-mean-square of the distance of molecular travel is given by :

$$X_{rms} = \sqrt{(2Dt_{recovery})} \tag{3}$$

Where D is the molecular diffusivity of the different molecules in the surrounding gas. In air, at 20°C and 1000 hPa, D is 0.16 cm^2.s^{-1} for CO_2, while for CH_4, it is 0.23 cm^2.s^{-1} (Massman, 1998). And $t_{recovery}$ is the waiting time before analysis

Then, during analysis, both molecular diffusion and Taylor dispersion affecting the sample have to be accounted for. During this phase the root-mean-square of the distance of molecular travel is given by :

$$X_{rms} = \sqrt{(2D_{eff}t_{analysis})} \tag{4}$$

Where $t_{analysis}$ is time needed for a parcel of the sampled air to reach the analyser's cell and D_{eff} is the effective diffusion coefficient combining molecular diffusion and Taylor dispersion given by:

$$D_{eff} = D + \frac{a^2\bar{V}^2}{48D} \tag{5}$$

where D is the molecular diffusivity of the different molecules in the surrounding gas, a is the tube's inner radius, and $\bar{V}$ is the average velocity of the air inside the tube.

In addition to the effects of diffusion and dispersion, which are the main drivers of the resulting vertical resolution, the smearing effect of the cell of the analyzer during analysis has to be taken into account. The analyzer used in this study (Picarro Cavity RingDown Spectrometer (CRDS) G2310) pulls the sample at 110 sccm and measuring at 0.5 Hz makes one measurement every 3.7 scc (standard cubic centimeters). The analyzer cell has a standard volume of approximately 6 scc, since it is 35 cc in volume, but is maintained at 187 hPa (140 torr) and 45°C. The volume of the cell needs to be flushed about 3 times for the air to be completely renewed (Stowasser et al., 2014).

To account for mixing in the volume of the cell of the analyzer, a Gaussian function characterized by the following standard deviation σ_{cell} is considered:

$$\sigma_{cell} = \frac{1}{2}\frac{V_{cell}}{\Sigma_{in}} = \frac{1}{2}l_{tube} \tag{6}$$

Where V_{cell} represents the volume of the cell, Σ_{in} the inner surface of a tube (πr_{in}^2), l_{tube} the AirCore length required to store the volume of the cell. A Gaussian function characterized by this standard deviation, allows to represent that mixing impacts a distance in the AirCore where almost 3 times the volume required to fill the cell is stored.

As all mixing effects can be considered Gaussian, the total distance of diffusion X_{rms} to be considered is given by:

$$X_{rms} = \sqrt{2Dt_{recovery} + 2D_{eff}t_{analysis} + \left(\frac{1}{2}l_{tube}\right)^2} \tag{7}$$

Using equation 7 and knowing that air is distributed in the AirCore as a linear function of total pressure column sampled, it is possible to evaluate the pressure range affected by mixing related to diffusion and dispersion. The factor 2 in front of X_{rms} comes from accounting for diffusion in both directions.

$$\Delta P = P_{max} \frac{2X_{rms}}{L} \tag{8}$$

5 Where ΔP represents the effective resolution and P_{max} the pressure at the surface when the coil is closed. L is the total length of the AirCore. In the case of 2 tubes or more, ΔP can be calculated independently for each tube.

Using a standard atmosphere temperature profile it is then possible using the hydrostatic law to associate the atmospheric pressure to a given altitude. In order to best represent the latitudes at which the AirCores are to be deployed, we used the average temperature profile of the representative TIGR (Thermodynamic Initial Guess Retrieval) dataset (Chédin et al. 1985, 10 available at http://ara.abct.lmd.polytechnique.fr/index.php?page=tigr) for mid-latitudes.

2.1.2 Aiming for a high resolution AirCore

To appreciate the value of the AirCore-HR it is important to understand the factors that determine the resolution of an AirCore. The first factor is the sample cell of the analyzer that will limit the number of independent measurements over the sampled volume. The second factor is the diffusion distance (explained above) which, depending on the diameter of the tube and the lag 15 between when air was sampled and when it is analyzed, will eventually limit the sampling resolution of the AirCore.

At mid latitudes, air sampled over a 10 hPa descent between 20 and 30 hPa represents about 3 km of vertical distance whereas air sampled over a 10 hPa descent between 450 and 460 hPa represents about 200 m of vertical distance. This has a direct consequence for the observation of the stratosphere, for which the sampled air needs to be preserved while sampling as much of it as possible. This can be achieved by combining sections of tubes of different diameters. A given volume of air 20 is indeed affected differently if stored in a section with a smaller diameter: although diffusion remains the same, the distance of travel for a molecule to impact an equivalent volume increases. Therefore, using at least 2 tubes, one characterized by a small diameter at the end that remains closed, and one characterized by a larger diameter at the end that remains open allows keeping a high resolution for the stratosphere (by storing the stratospheric part of the sampled profile in the tube with the smallest diameter), while still sampling a consequent volume of air thanks to the larger tube. To maximize the total volume 25 of the AirCore-HR and limit the impact of the diffusion distance the AirCore-HR was designed with tubes of two different diameters.

Figure 2 illustrates a comparison of the vertical resolution of CO_2 measurements that can be expected for air sampled with different AirCores (for an analysis performed at 38.5 sccm, with a surface pressure Pmax of 1013.25 hPa and a given storage time of 3 hours). The resolution achievable with the first AirCore designed by NOAA (Karion et al., 2010) is shown in black. 30 After 3 hours of waiting time before analysis, it is possible to achieve a vertical resolution of 250 m at 10 km and 1,2 km at 20 km.

In order to achieve a higher resolution along the whole atmospheric column, a design of a 300 m tube consisting of a 200 m of 1/8 in.(3.175 mm) tube and a 100 m of 1/4 in.(6.35 mm) tube linked together as one tube was selected for AirCore-HR.

The increase in overall volume of the AirCore-HR allows a significant increase in resolution throughout the whole sampled air column (Fig. 2) albeit an increase of the overall weight. The resolution of the AirCore-HR for CO_2 is estimated to be better than 300 m up to 15 km and better than 500 m up to 22 km.

The resolution achievable by the light-weight AirCore-GUF designed and developed at Goethe University Frankfurt is also shown in Fig. 2. AirCore-GUF is a 100 m long combining 3 tubes: a 20 m-long 8 mm tube, a 40 m-long 4 mm tube and a 40 mm-long 2 mm tube. It has been designed to be carried by meteorological balloons, making compromises between weight and achievable resolution. Thanks to a third tube with thinner diameter, it enables a good resolution in the stratosphere (700 m at 20 km).

The storage time between landing and analysis of the sample is a key factor influencing the vertical resolution. The resolutions plotted in Fig. 2 are for a storage time of 3 hours. Figure 3 shows the degradation of the resolution for various storage times of an air sample captured by the AirCore-HR. While a 6-hr delay (blue plot) will still preserve a resolution better than 300 m up to 20 km, waiting for 12-hr after landing to perform the analysis will reduce the resolution to 500 m at 20 km (green plot). The impact of the delay on the achievable vertical resolution with the sampled profile is also presented for a 24-hour delay (orange) and a week delay (red). It is very clear from this figure that to avoid loosing vertical resolution one of the challenges of using AirCores is to quickly recover the sample and perform the analysis.

2.2 AirCore-HR experimental setup

In order to obtain the vertical resolution shown in Fig. 2, the AirCore-HR comprises two tubes linked together as one, yielding an overall length of 300 m, a weight of 14.3 kg and an inner volume of 2.967 L. The detailed characteristics are given in Table 1. Both tubes have been treated by Restek, Inc., with Sulfinert® coating to reduce interactions of the sample with the walls.

The overall design is plotted in Fig. 4. Both sides of the coil are connected to 3-way valves that allow ambient air to flow either through the AirCore-HR or through a bypass. This bypass consists in a 1 m long, 1/16 in. (1.5875 mm) diameter stainless steel tube that allows pulling air into the analyzer bypassing the AirCore-HR. During flight, in addition to this setting, a dryer consisting of a short length (10 cm) of stainless steel tube filled with fresh magnesium perchlorate is positioned at the open end of the tube (at the entrance of ambient air on the solenoid valve) to ensure that no moisture enters the tubes during sampling. The additional volume to the system is very small and represents less than 0.005 % of the total volume (it is thus not considered in the calculation the vertical resolution (sect. 2.1.1)).

The AirCore-HR payload has been designed to fit into a polystyrene foam box. It is flown together with an electronic data package designed at LMD that collects meteorological data from a pressure sensor and 3 temperature probes and also controls the opening and closing of a solenoid valve at the open end of the AirCore. Temperature probes are placed along the AirCore in contact with various segments of the tube and allow monitoring the mean temperature along the coil during the flight. The pressure sensor is an absolute pressure sensor that measures the ambient air pressure during the flight.

2.2.1 Laboratory testing

Several tests were conducted in the laboratory under monitored conditions to evaluate the overall consistency of the AirCore-HR. In particular, the AirCore-HR has been tested for leaks at the junctions, and at the valves used as closing points on each side of the AirCores. To test the preservation of the concentration of the sample, calibrated dry standard gases of two different values for both CO_2 and CH_4 are used. In repeated experiments under various conditions, it was noted that to reach the optimal Picarro CRDS precision at 0.5 Hz of 0.07 ppm for CO_2 and 0.5 ppb for CH_4 the tubes conducting the sampled air have to be perfectly dry. Therefore, the analysis line is systematically dried out by flowing dry air through the bypass. Water vapor concentration is closely monitored with the Picarro to be lower than 0.002 % before conducting any tests with AirCore-HR. This method allows to eliminate significant biases and to obtain mean measurements.

2.2.2 Atmospheric gas standards

For testing and analysis of the AirCores, two calibrated gas standards are used. The cylinders are connected to a multiport valve allowing selection of one of the gases.

The first standard is composed of high concentration CO_2 and CH_4 of respectively about 400 ppm and 1900 ppb and referred to as 'high concentration calibration standard'. The other standard is composed of lower concentration of CO_2 and CH_4 of about 360 ppm and 1700 ppb respectively and referred to as 'low concentration standard'. The gas cylinders have been calibrated on WMO scales at Laboratoire de Sciences du Climat et de l'Environnement (LSCE) (courtesy of Michel Ramonet and Marc Delmotte) and the exact calibrated values of the standard can be found in Tab. 2 where CO_2 concentrations are given on the WMOX2007 scale and CH_4 concentrations are given on the NOAA-2004 scale.

2.2.3 The Picarro CRDS Analyzer

All gas analyses of LMD AirCores were performed using one trace gas analyzer by Picarro, Inc., model G2310 (Crosson, 2008). The analyzer tightly controls the pressure and temperature in its measurement cell [187 hPa (140 torr) and 45°C], to achieve the above precision (see Sect.2.2.1). The sample flow rate was controlled by a critical orifice placed at the outlet, limiting the flow at 38.5 sccm during analysis.

2.3 Processing method

Upon recovery, the AirCore-HR is plugged into the prepared analysis system. It is first kept closed on both ends, allowing to pull calibrated standards through the bypass into the analyzer. Once the values measured with the continuous analyzer are stabilized to the expected values for the calibration standard used as 'push gas', the analysis of the air captured in the coil can start. This phase is very important to make sure that, after plugging the AirCore-HR in the system, the mixing ratio read by the Picarro is not contaminated by water vapor that can have entered the analysis chain. The collected sample is then analyzed by opening both ends simultaneously; the air is pulled from one end into the continuous analyzer and low concentration calibration standard is pulled through the other end. The top of the profile with the remaining fill gas is pulled first into the analyzer.

The calibrated gas standards given in Tab. 2 allow replacing the values read by the Picarro onto the WMO scale. The high concentration standard is used as fill gas to have a noticeable difference between fill gas and stratospheric air sample at the top of the profile. The low concentration calibration standard is chosen to be used as push gas to have a noticeable difference of the mixing ratios compared with the expected values of CO_2 and CH_4 at the surface.

5   Several steps are required to accurately place the Picarro measurements on a vertical scale in order to retrieve the vertical profiles. The dry mole fraction of CO_2 and CH_4 provided by the Picarro are used. These are automatically corrected by the instrument for a combined effect of dilution and line broadening caused by water vapor (Chen et al., 2010; Rella et al., 2013). Then, in a first processing step, the measured mixing ratios are corrected for a bias from the Picarro measurement to the WMO scales. The correction is calculated thanks to the measurement of the calibrated standards by the Picarro at the time of analysis.

10   Figure 5 shows an example of CO_2 and CH_4 mixing ratios measured by the Picarro instrument during the StratoScience 2014 campaign. In order to extract the measurements corresponding to the sampled air, the top and the bottom of the profiles need to be defined. The top of the AirCore sample is considered to be at midpoint of the transition in concentration between the push gas and the remaining fill gas. This point corresponding to an estimated pressure of 0 hPa in the tube is marked with a green cross in Fig. 5. The bottom of the profile is defined at midpoint on the transition of concentration between 'push gas'

15   and sampled air. It is marked with a red cross in Fig. 5.

As a first approach, it is assumed that the air entering the tube equilibrates the sample with ambient pressure and adjusts very quickly with the mean coil temperature. As the characteristics of the AirCore (length, diameter) do not change, ambient pressure and mean coil temperature are the two main factors that regulate the number of moles in the AirCore. Using the ideal gas law (equation 9), it is possible to calculate the number of moles captured in the tube all along the trajectory.

$$20 \quad PV = nRT \Leftrightarrow n = \frac{PV}{RT} \tag{9}$$

where P is the ambient pressure, V the inner volume of the AirCore, n the fraction of moles, R is the universal gas constant in $J.K^{-1}.mol^{-1}$ and T the ambient temperature in Kelvin.

With measured time-series of pressure (P_i) and temperature (T_i) (Fig. 7), it is possible to relate the number of air moles in the tube (n_i) to the atmospheric pressure at any given time during the flight:

$$25 \quad n_i = \frac{P_i V}{RT_i} \tag{10}$$

This number is maximum when the AirCore reaches the Earth's surface, i.e :

$$n^{max} = \frac{P_s V}{RT_s} \tag{11}$$

Where P_s and T_s correspond to the surface pressure and to the temperature of the AirCore when landing at the surface.

The critical orifice setting the flow during analysis at 38.5 sccm/min and the controlled settings of the Picarro analysis cell

30   ensure that the same number of moles are analyzed at every time step. In other words, during the sample analysis, the number

of moles flown through the analyzer increases linearly with time. Hence, the number of moles at any time during the analysis is

$$n_i = n^{max} \frac{t_i}{\Delta t} \tag{12}$$

where Δt is the total time duration of the analysis between the defined top and bottom of AirCore sample.

Using equations 10 and 12, a specific pressure point can be associated to every Picarro measurement of the sample to retrieve the vertical profiles. Although the process is quite simple, the selections of start and end point of the sampled profile in the Picarro data as well as in the temperature and pressure data are delicate steps that have a direct impact on the resulting profiles (see Section 4.2). Two additional effects need to be taken into account: the pressure loss along the tube (P. Tans, pers. comm.), and the accounting of potential losses of air samples during the recovery process (see Section 3.2).

3 The StratoScience 2014 campaign

3.1 The EDS-Stratéole flight

AirCore-HR was flown for the first time during the Stratoscience campaign operated by the French space agency (CNES) in collaboration with the Canadian Space Agency (CSA) in Timmins (Ontario, Canada, 48.57 North -81.36 East) in August 2014. It participated in the third flight of the campaign: the "EdS (Effet de Serre – Greenhouse Effect) - Stratéole" flight.

The carrier consists of a gondola that could accommodate a total of 8 instruments including the AirCore-HR. All these instruments (consisting of small packages of several kg) were brought together on the same structure with the aim of studying simultaneously several climate variables. In total, the Gondola weighed 248 kg.

In addition to AirCore-HR, two AirCores-GUF from Goethe University Frankfurt were also flown during this flight.

3.2 Flight trajectory

To fulfill the requirements of the 8 instruments, the EDS-Stratéole flight had a very specific flight trajectory. The takeoff (release of the balloon) took place on the 28th of August 2014 at 20:33 local time in Timmins (00:33, 29/08/2014 UTC). After the ascent phase, the flight consisted in a monitored and controlled descent with two stops. Following a short stop at the ceiling at a barometric altitude of 14 hPa (29km), an evacuation trap allowed to let some gas out to engage in a descent phase down to a barometric altitude of 54 hPa ($\approx$20 km); the balloon then stabilized in a slow descent phase during 6 hours down to the barometric altitude of 78 hPa ($\approx$18 km). The separation between the flight chain and the balloon did not take place at the ceiling as for weather balloon flights but at the end of this slow descent at a barometric altitude of 78 hPa. The two elements (flight chain and balloon envelope) were then separated and engaged separately in faster descent, finally both landed in a dry area 350 km South-East from Timmins at 7:28 local time (11:28, 29/08/2014 UTC).

Figure 6 shows the flight profile together with the main operating states of AirCore-HR and AirCore-GUF. First, during a preparation phase on the ground before flight (marked in blue), the AirCore-HR was filled with the high concentration

calibration standard chosen as 'fill gas'. Then, the AirCore-HR was placed on the gondola and opened on one end just before takeoff. During the ascent phase (marked in green) the AirCore-HR emptied as it equilibrated with ambient pressure, thus evacuating fill gas. To preserve some part of the initial fill gas in the coil the AirCore-HR was closed at 19h Pa ($\approx$27 km) through a signal sent to the solenoid valve. The AirCore-HR remained closed at ceiling (marked in red) and was then re-opened by sending another signal to the solenoid valve at 19 hPa during the descent phase. During all the descent phase (marked in black), the AirCore-HR remained open at one end. As the coil equilibrated with ambient pressure, air was pulled into the tube. At landing, after the pressure sensors on the electronic package detected no more pressure change, the solenoid valve closed in order to preserve the sample while waiting for recovery.

Joint efforts of CNES and CSA teams allowed accessing the AirCores and performing the analysis less than 3 hours after landing. Unfortunately, at the end of the flight, the electronic circuit maintaining the solenoid valve closed experienced a short power cut of about an hour which resulted in sampled air evacuating from the AirCore. The AirCore-HR coil heated up after reaching the ground and since it had been exposed to cold temperatures during the flight. During this period, the heating that occurred resulted in losing a fraction of the profile equivalent to the air sampled from 900 to 980 hPa. The loss of that fraction of the total sample had an impact on the retrieved vertical profiles (see Section 4.1).

The specific periods of interaction with ambient air of the AirCores-GUF are highlighted in Fig. 6 (b). The main difference between AirCore-HR and AirCore-GUF was the lack of a closing device for the latter. The Light AirCores from Goethe University Frankfurt thus remained open until recovery. Being less insulated than AirCore-HR and exposed to the same cold temperatures during flight, AirCores-GUF lost a fraction of the profile equivalent to the air sampled from about 780 to 980 hPa.

3.3   Measurement of additional data

In order to determine the vertical profiles of CO_2 and CH_4 from the analysis of sampled air, measurements of several atmospheric parameters are needed (see Section **2.3**). The two most important parameters are the ambient pressure and the mean coil temperature. Those parameters are recorded by the AirCore-HR electronic data package (Section 2.2). Figure 7 shows the evolution of coil temperature and ambient pressure measured during the flight. The temperatures recorded by the 3 temperature probes during flight are plotted in red (full, dashed, points) and reported on the right y-axis. Mean coil temperature is obtained by taking the mean of 3 temperatures recorded by independent probes located at different positions along the AirCore-HR. The ambient pressure during the flight is plotted in black and reported on the left y-axis.

Comparison between AirCore-HR and other pressure measurements highlighted a small drift in AirCore-HR data pressure recordings. Therefore the pressure profile recorded with the electronic of AirCore-HR has been corrected to fit the high precision of the records of a Paroscientific. Inc.© absolute pressure gauge which is characterized by an accuracy of 10 Pa and a precision of 0.1 Pa.

Additionally, GPS coordinates and altitudes from CNES were used to complete the dataset.

4 Results: The 0-25 km CO_2 and CH_4 profiles

4.1 The profiles

Figures 8 (a) and (b) show the CO_2 and CH_4 profiles measured during the StratoScience 2014 campaign. Each profile comprises about 1800 points on the vertical. As explained in Section **3.2**, profiles stop at 900 hPa due to the sample loss after landing. Both CO_2 and CH_4 AirCore-HR vertical profiles reveal thin structures of the atmosphere and airmass transport signatures. Figure 8 (c) shows the ambient temperature recorded onboard during flight. From this ambient temperature profile the tropopause was estimated to be at 162.1 hPa according to the definition of the WMO thermal tropopause (Reichler et al., 2003).

In Fig. 8 (a), a strong decrease of CO_2 can be observed in the first layers above ground. This is coherent with CO_2 uptake by vegetation near the surface during the summer season. CO_2 then reaches higher values in the free troposphere ($\sim$393 ppm), with small variations (of 0.5-2 ppm) albeit 2 well-marked signatures at 700 and 600 hPa. CO_2 reaches its highest value of 396 ppm just above the tropopause. In the stratosphere, CO_2 values are expected to be lower since the exchange rate between upper troposphere and lower stratosphere takes several years (Boering et al., 1981; Andrews et al., 2001; Engel et al., 2002). Above $\sim$110 hPa, CO_2 mixing ratios decrease slowly from 396 ppm to 385 ppm at 30 hPa albeit one structure captured at the very top of the profile between 30 and 40 hPa. This structure is correlated with a similar one at the same barometric altitude in the CH_4 profile in Fig.8 (b).

As can be seen in Fig. 8 (a), part of the CO_2 profile between 70 and 90 hPa is shown in dotted line. This corresponds to the range of the 7-hour plateau phase (cf. Fig. 6) where unrealistically low CO_2 values were sampled (visible in the CO_2 data in Fig. 5 at the analysis times between 300 and 450 seconds). The origin of these very low values of CO_2 is still debated. The alteration of CO_2 sampled in this range was possibly caused by the dryer. Indeed, the magnesium perchlorate used as drying agent at the entry point of the tube is slightly reactive with CO_2 and inert with CH_4. Because of the long exposure ($\sim$7h) to this drying agent during the plateau phase, enhanced by air coming in and out of the tube because of oscillations of the gondola around 80 hPa, the magnesium perchlorate might have impacted CO_2 sampled by the AirCore-HR. The dryer being inert to CH_4, no impact is seen on CH_4 profile. Other explanations might be that the air sampled during this particular phase was polluted through interaction with polystyrene, with the balloon envelope (pumping up some of the ambient air as they re-equilibrate with ambient air during this long phase) or by chemical interaction with helium from the balloon (during some short re-ascent phases).

The CH_4 vertical profile is presented in Fig. 8 (b). Mixing ratios of CH_4 have a small variability in the troposphere between 1800 ppb and 1880 ppb. The zoom on the tropospheric part (between 200 hPa and 1000 hPa) reveals pronounced structures captured in the troposphere, particularly in the region from 200 hPa to 700 hPa. These could be caused by transport or variability in the emissions. The strong decrease of CH_4 in the stratosphere is particularly well seen in Fig. 8 (b), with values of $\sim$1800 ppb near the tropopause at 120 hPa to 1100 ppb at 30 hPa. Along the slopes, several structures can be identified around 80 hPa and between 30 and 40 hPa, revealing transport patterns in the stratosphere.

A comparison between Fig. 8 (a) and (b) shows CO_2 variability is higher near the ground, whereas CH_4 variability is higher in the mid-to-upper troposphere and in the stratosphere. This is in agreement with the fact that CO_2 may have negative and

positive anomalies at the surface (associated mainly with vegetation uptake and anthropogenic emissions) whereas CH_4 has mostly positive anomalies coming from the surface and negative anomalies coming from the stratosphere.

A comparison was performed with CO_2 and CH_4 forecasts from the Copernicus Atmosphere Monitoring Service (CAMS) using the European Centre for Medium-range Weather Forecasts (ECMWF) model (Agusti-Panareda et al., 2014; Massart et al., 2014). This comparison is presented in Fig. 9. The tracer transport in the forecast is constrained with meteorological observations by re-initializing the forecast every 24 hours with operational ECMWF analyses, whereas the atmospheric CO_2 and CH_4 tracers are cycled from one 1-day forecast to the next, as in a free run. Therefore, the forecast is essentially a model simulation with state-of-the-art representation of tracer transport available in forecast mode (http://macc.copernicus-atmosphere. eu/d/services/gac/nrt/rt_fields_ghg). The CAMS-ECMWF CO_2 and CH_4 forecasts used here have a horizontal resolution of around 16 km x 16 km and a vertical resolution of 137 levels from the surface to 0.01 hPa. These forecasts have been collocated in space and time with AirCore-HR landing coordinates. 24 ppb were added to the CAMS-ECMWF CH_4 high resolution forecast to emphasize the good agreement on structures, rather than focusing on the bias which may be attributed to incorrect surface fluxes or issues with airmass exchanges along the vertical.

The agreement between both CO_2 profiles (Fig. 9 (a)) is satisfying throughout the troposphere in terms of structures. The forecast correctly reproduces the strong decrease in CO_2 from 800 hPa to the surface, as well as the increase in concentration from 800 hPa to 600 hPa, and a lower increase from 600 hPa. In the upper troposphere, from 300 hPa up to the tropopause at 150 hPa, the forecast displays different structures than those measured by the AirCore-HR. In the lower stratosphere (from 150 hPa to 90 hPa), the AirCore-HR and the CAMS-ECMWF forecasts both reveal a decrease in CO_2 starting from just above the tropopause up to the top of the stratosphere.

Although fewer vertical structures are seen in the forecast, the CH_4 mixing ratios and position of the broader vertical structures fit quite well with the measurements up to 200 hPa (Fig. 9 (b)). For lower pressures, the decrease of CH_4 measured by AirCore-HR is much more pronounced than the one simulated by the forecast. This is a known problem in the CAMS-ECMWF model which is currently being investigated (A. Agusti-Panareda, S. Massart, pers. comm.) and that was also discussed in Verma et al. (2016).

4.2 Associated uncertainties

Monte Carlo simulations were performed to assess the uncertainty associated with the retrieved constituent profiles. The retrieval process of the vertical profiles was iterated a 1000 times by randomly changing the original datasets within the estimated uncertainty range of every identified uncertainty source. This allowed to produce a set of 1000 slightly different outcomes for the vertical profiles both in terms of mixing ratios and vertical position. A standard deviation of the mixing ratios at a given position was then calculated based on this dataset. In these simulations we took into account the following uncertainties :

(i) the accuracy of the gas analyzer: Picarro measurement accuracy was defined as a Gaussian standard deviation of the mixing ratios based on the instrument specification (i.e deviations of 0.5 ppb for CH_4 and 0.07 ppm for CO_2 (Crosson, 2008)).

(ii) the mean temperature profile: To account for the impact of temperature correction, the temperature profile was randomly chosen among the 3 profiles measured by the 3 probes. Indeed, the 3 temperature probes are placed at different positions along

the tube (near the entrance, in the middle of the AirCore and near the closed end) and depending on the distance to the inlet they have recorded different temperatures along the AirCore. Choosing randomly between one of the 3 probes is thus the conservative way accounting for the uncertainty related to the mean temperature of the AirCore.

(iii) the pressure profile : an uncertainty of 0.1 Pa corresponding to the accuracy of the Paroscientific. Inc.© absolute pressure
5   gauge was used.

(iv) the selection of the sample: The choice of the exact midpoint of transition between either push-gas and sample, or remaining gas and sample (see Section **2.3**) has an impact on the altitude of both ends of the profile. A random uncertainty of $\pm$ 1 Picarro measurement point was defined for both uncertainties.

(v) the potential loss of air sample resulting from the tube remaining open after landing as occurred during this flight (see
10   Sect. **3.2**): An uncertainty of $\pm$ 10 hPa was associated to the bottom pressure correction that was defined to take the air loss into account.

The uncertainties discussed here are related to the analysis and processing of the sampled air and are only valid for the AirCore-HR in the case of this flight and may have different results in other situations. The CO_2 uncertainties do not take into account the potential interactions with the drying agent that are hard to quantify and need to be further studied (cf Section 4.3).
15   The uncertainties associated to the AirCore-HR CO_2 and CH_4 profiles were calculated for the 5 uncertainty sources together but also separately to estimate the influence of each individual source of uncertainty. The overall resulting uncertainties associated to the mixing ratios of CO_2 are presented in Fig. 10 and those associated to the CH_4 mixing ratios are presented in Fig. 11. Figures 10 (a) and 11 (a) show a reminder of the vertical profiles of CO_2 or CH_4 respectively. Figures 10 (b) and 11 (b) show the uncertainties without taking the uncertainty source (v) into account in order to illustrate what the expected uncertainties
20   would be in an ideal case where the closing system would have worked. Figures 10 (c) and 11 (c) show the overall uncertainties associated to the vertical profiles accounting for all uncertainties sources previously listed.

Comparing Fig. 10 (b) with Fig. 10 (c) and Fig. 11 (b) with Fig. 11 (c) shows that, for both CO_2 and CH_4, the uncertainty related to the bottom pressure correction has an important impact on the uncertainties estimated in the troposphere although in the stratosphere the uncertainties remain relatively not impacted by this. Indeed the fraction of the overall uncertainties (Fig. 10
25   (c) and Fig. 11 (c)) that is related to the loss of air is above 80% in the troposphere and drops to about 30% in the stratosphere. The dominating uncertainty source in the stratosphere is related to the selection of the sample. Mis-selecting the transition point between the gas in the AirCore sample and the calibrated standard by only one measurement has an important impact on the positioning of the stratospheric part of the profiles. Indeed, the whole stratospheric air sampled by the AirCore accounts for about 8% of the total sample ($\sim$150 points out of $\sim$1800 total points), but corresponds to $\approx$ 15 km of the 25 km profile.
30   Hence, a difference of a single measurement point in the positioning of the profile does matter.

Additionally, the impact of the variability in the measurements of the 3 temperature probes has been studied. It was found that temperature uncertainty has a very limited influence on the overall uncertainties, of the order of 6 %, despite differences of several °C (Fig. 7). Although differences up to several °C are observed between the measurement, the overall variation of the temperature is captured similarly by the 3 temperature probes (Fig. 7). The increase of sampled moles in the AirCore at
35   each pressure level as well as the total number of sampled moles in the AirCore are almost unchanged when considering one

or the other temperature sensor. This comes from the fact that, during the fast descent phase in the troposphere when most of the sampled air is captured the temperature remains very stable.

Overall, the average uncertainty on the CO_2 profile (Fig. 10 (c)) is 0.24 ppm throughout the column. The average uncertainty in the troposphere is 0.25 ppm with relatively higher uncertainties in the bottom of the profile where important variations of CO_2 are measured, inducing that the slightest positioning uncertainty translates into mixing ratio uncertainties. For the whole stratosphere above 120 hPa, where the CO_2 profile is more stable, the average uncertainty drops to 0.11 ppm.

The average uncertainty on the overall CH_4 profile (Fig. 11 (c)) is 2.78 ppb. In the stratosphere, above the tropopause at 120 hPa, CH_4 uncertainties are quite variable along the profile, and can be as high as 10 ppb locally but on average are estimated to be 6.42 ppb. Such high values stem directly from high vertical gradients in mixing ratios: in that case, the assumed error on the vertical positioning of the profiles translates into higher uncertainties. In the troposphere, the average uncertainty for the CH_4 profile is below 2 ppb with sometimes values up to 5 ppb where the vertical profile shows transport structures of 30 ppb or more along the vertical in the troposphere.

4.3 Comparison between AirCores with different resolutions

4.3.1 Overall comparison

Benefiting from the accommodation of several AirCores on-board the CNES gondola, the AirCore-HR profiles can be compared with those of the lighter AirCores-GUF (see Section 3). AirCore-GUF air samples were processed at LMD using the same methodology as for AirCore-HR (Section 2.3). The processing took into account the fact that AirCores-GUF remained open for 3 hours before being manually closed at recovery leading to the loss of the bottom of the profile between 980 and 780 hPa (see Section 3.2). Both AirCore-GUF being identical, the comparison is presented with only one AirCore-GUF in order to focus on the comparison between AirCores with different resolutions. Figures 12 (a) and (b) show the comparison of AirCore-HR and one AirCore-GUF CO_2 and CH_4 profiles.

The particular descent profile of this flight had several impacts on the AirCore-GUF profiles:

(i) as for AirCore-HR, unrealistic low values of CO_2 were sampled during the long plateau phase that happened between 70 and 90 hPa;

(ii) since the lower resolution AirCore-GUF captured a smaller volume than AirCore-HR, the stratospheric part of the profile was impacted by diffusion during the 7 hours plateau phase. Indeed, during the plateau phase at about 90 hPa, the air sampled from 20-90 hPa by AirCore-GUF remained in the first tube of 20 m / 8 mm diameter whereas it was stored over the 100 m / 1/4 in.(6.35 mm) tube for AirCore-HR. This lead to a more intense diffusion in the AirCore-GUF sample.

Therefore, all the CO_2 sampled above 100 hPa with AirCore-GUF was probably altered by the combination of unrealistic low values of CO_2 acquired during the plateau phase and the diffusion effects. This part of the profile that should not be considered is shown in dotted line as with AirCore-HR CO_2. Only AirCore-GUF CO_2 sampled in the troposphere below 100 hPa should be compared, where residual effects from this phase are minimal. Concerning CH_4, which was not impacted by

the dryer during the plateau phase, only the difficulty of properly modeling the diffusion inside the tube remains. The full AirCore-GUF CH_4 profile is shown but the stratospheric part of the profile should thus be taken with caution.

The comparison between AirCore-HR (black) and AirCore-GUF (blue) highlights that both CO_2 profiles (Fig. 12 (a)) have a good agreement in terms of structures. In particular, the impact of vertical resolution is well seen in Fig. 12 (a), with less

5 structures captured by the lower resolution AirCore. However, there is a variable but noticeable bias between the profiles (up to 3 ppm in some part of the profiles).

For CH_4, Fig. 12 (b) reveals that the agreement is excellent between AirCore-HR and AirCore-GUF. The zoom on the tropospheric part (between 200 hPa and 1000 hPa) shows that the different AirCores capture the same structures and allow to retrieve similar vertical profiles both in terms of structures and mixing ratios albeit at a different resolution. In the stratosphere,

10 both AirCores capture the position and inclination of the decreasing slope of methane. However, some stronger differences can be seen in terms of mixing ratios between both profiles between 70 and 90 hPa or between 30 and 40 hPa. In these ranges, similar structures are captured by both AirCores but seem to be very strongly impacted by diffusion in the AirCore-GUF CH_4 profile. This illustrates the impact of diffusion, which is stronger for AirCore-GUF than for AirCore-HR during the long plateau phase.

15 Overall, the comparison between both AirCores reveals that the high resolution captures more information on the vertical distribution along the atmospheric column.

4.3.2 Degradation of the resolution

To perform a fair comparison between the different AirCore profiles, the degradation of the resolution of AirCore-HR profiles to that of lower resolution AirCore-GUF has to be performed. This exercise aims also at evaluating the theoretical calculation

20 of the expected resolution (Section. 2.1).

The vertical resolutions shown in Fig. 2 were calculated for a standard atmosphere and air sampled from 10 hPa to a ground pressure of 1013.25 hPa. In order to account for the sampling that occurred during flight and how the sampled air was distributed within the tubes, the vertical resolution of AirCores-GUF was recalculated with the specific parameters of the flight for both CO_2 and CH_4.

25 Degradation of the AirCore-HR profiles is performed through the convolution with a Gaussian window with a standard deviation of the lower vertical resolution at each given altitude:

$$g(x) = \frac{1}{\sigma\sqrt{2\pi}}\exp(-\frac{x^2}{2\sigma^2}) \qquad (13)$$

where σ is the standard deviation (i.e the vertical resolution) at a given vertical position x.

It allows retrieving a degraded version of the profiles :

30 $$degradedX_{CH_4}(x_0) = \int CH_4(x)g(x-x_0)dx \qquad (14)$$

The degraded version of the CO_2 profile is calculated similarly. To avoid the parts of the profiles that may have been affected by the strong diffusion during the long plateau phase in the flight profile, the comparison with degraded AirCore-HR profiles is only presented for pressures higher than 200 hPa.

The effect of the degradation of the AirCore-HR profile to the lower resolution of AirCore-GUF is presented in Fig. 13 (a) and Fig. 14 (a). The differences between AirCore-GUF and the smoothed version of AirCore-HR (degraded to the vertical resolution of AirCore-GUF) are shown in Fig. 13 (b) and Fig. 14 (b).

The comparison of the the CO_2 profiles in Fig. 13 (a) shows that when the lower vertical resolution of AirCore-GUF is taken into account, both AirCores display very similar structures. However, a bias remains between both profiles. The difference between AirCore-GUF and the smoothed AirCore-HR (Fig. 13 (b)) highlights that this bias varies linearly from -1 ppm at 200 hPa up to 3 ppm at 780 hPa. The reasons of these observed difference are still debated. The main hypotheses are that it could be related to some kind of ''memory effect' of the tubing to the previously stored calibrated gas or it is also suspected that the individual dryers from different AirCores may affect the CO_2 samples slightly differently when capturing CO_2. Overall, the problem highlights that there are some remaining questions with CO_2 sampling and that some potential interferences with CO_2 have to be studied more closely.

Concerning CH_4, the degraded AirCore-HR profile (pink) smears out the smaller structures that were captured by the high resolution and matches perfectly the AirCore-GUF profile both in terms of structures and mixing ratios. The difference between AirCore-GUF (blue) and the smoothed AirCore-HR profile (pink) vary between -2.7 and + 2.1 ppb (Fig. 14 (b)) which is in agreement with the 2.8 ppb average uncertainty that can be associated to the AirCore-HR profile (cf sect. 4.2). In addition to allowing the comparison of AirCore-HR profiles with those of lower resolution AirCores, the excellent agreement of both CH_4 profiles validates the computation of theoretical vertical resolution presented in Section **2.1.2**.

5   Conclusions

In this paper, a new AirCore (AirCore-HR) allowing high resolution measurements of CO_2 and CH_4 from the ground up to almost 30 km is presented. Thanks to the combination of 2 tubes, it allows retaining air samples with a vertical resolution better than 500 m up to 20 km when the analysis is performed within 6 hours after landing of the instruments. As for any AirCore, the final resolution depends on the delay between landing and analysis.

The AirCore-HR was flown for the first time on a multi-instrument gondola which allowed to perform comparisons of the vertical profiles retrieved with AirCore-HR and lower resolution AirCore-GUF. The degradation of the profile given by AirCore-HR to the resolution of AirCore-GUF revealed an excellent agreement between both profiles for CH_4 which fully validates the theory behind AirCores.

CO_2 profiles retrieved from the AirCores on this flight have revealed unexpected structures between 60 and 90 hPa when the flight experienced a long plateau phase of about 7 hours during descent, not seen on CH_4. It is suspected that the magnesium perchlorate used as drying agent at the inlet of the AirCores inert to CH_4 may have played a role in the alteration of CO_2 during this particular phase at low pressure. Moreover the comparison of CO_2 profiles has highlighted that the agreement is

good in terms of structures, but that an important and variable bias is seen between profiles. This bias is also suspected to come from potential interaction with the dryer and shows that CO_2 sampling aspects with AirCores as well as these potential impacts of the drying agent have to be further studied. Therefor, specific tests are planned during the future Strato-Sciences 2017 campaign that will take place in March-April 2017 in Alice Springs, Australia. These tests will include comparing several

5   independent AirCores flown on the same Gondola with and without a dryer at inlet.

By designing a method taking into account all the sources of uncertainties in the processing of the data, the overall uncertainty is estimated to be less than 3 ppb on the CH_4 profile and less then 0.3 ppm on the CO_2 profile. A particular issue during the flight with the closing system has led to the loss of part of the sampled air. Therefore the highest pressure point sampled by the AirCore-HR had to be corrected. An uncertainty of 10 hPa was associated to this correction and it was estimated that this

10   uncertainty is responsible for $\sim$80% of the overall uncertainty on the profiles. In an ideal case where the system would close and retain the complete sample, it would be possible to know more precisely the pressure at which air was sampled last and thus to improve the overall uncertainty to about 0.1 ppm for CO_2 and 2 ppb for CH_4.

Comparison between AirCore data and forecasts from CAMS-ECMWF has yield satisfying agreements between AirCore-HR profiles and simulated profiles. In particular, well pronounced vertical transport signatures in the troposphere in both CO_2

15   and CH_4 profiles are similar for both the forecasts and AirCore-HR profiles. In the stratosphere, AirCore-HR CH_4 profile seems to indicate that the decrease of stratospheric CH_4 in the forecasts is too slow, which may have an important impact when deriving total or partial columns of CH_4 from the analyses.

This comparison illustrates the potential of AirCores to evaluate atmospheric transport models, as well as GHG satellite retrievals from TIR and SWIR instruments. Especially, light AirCores flown from weather balloons could be deployed at

20   various locations to complete an effective system together with ground stations and regular aircraft campaigns. Such lightweight systems could also contribute to specific campaigns for calibration and validation of future space missions. In order to fit these applications, the spatial and temporal resolution requirements necessary to evaluate the models or satellite retrievals efficiently need to be assessed.

Along with the development of robust lightweight systems, it is also important to continue development strategies of Air-

25   Cores for large platforms carrying heavy payloads. Such platforms, flown during specific stratospheric balloon campaigns, allow unique multi-instrument measurements of the same or complementary atmospheric variables. The simultaneous use of laser-diode spectrometers, cryosamplers and AirCores, that can only be performed during these specific campaigns are necessary to evaluate the retrievals performed with various AirCores and test improvements of the instruments.

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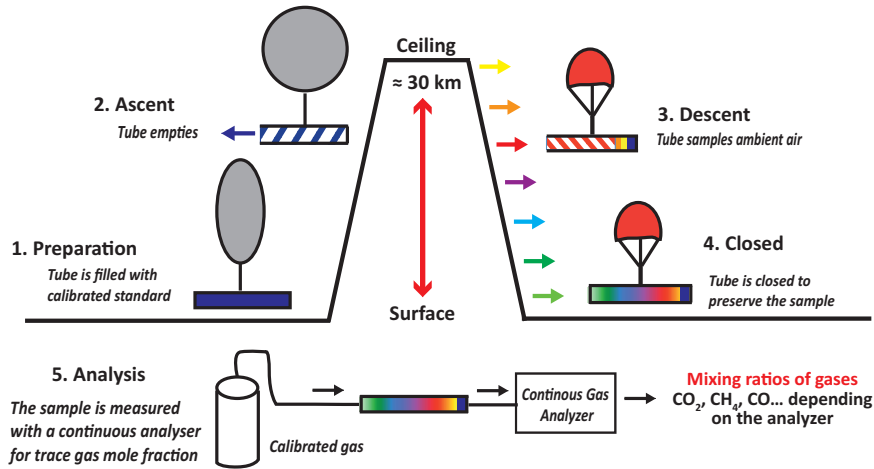
Figure 1. Schematic description of the 5 steps of the AirCore sampling method.
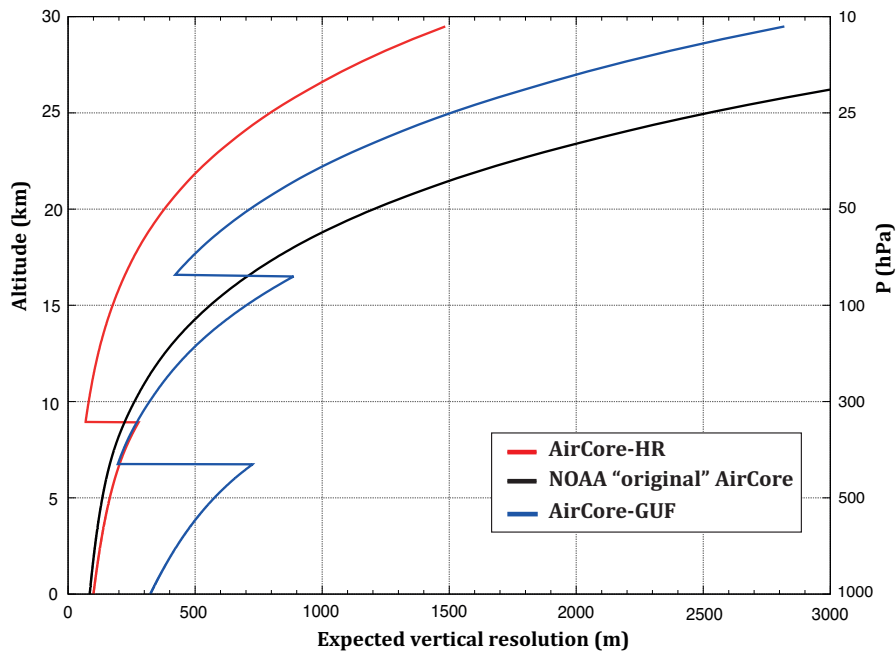
Figure 2. Comparison of the vertical resolution that can be expected with different AirCores for CO_2 measurements after 3 hours storage time before analysis. AirCore-HR (red), the original NOAA AirCore (black), AirCore-GUF (blue).
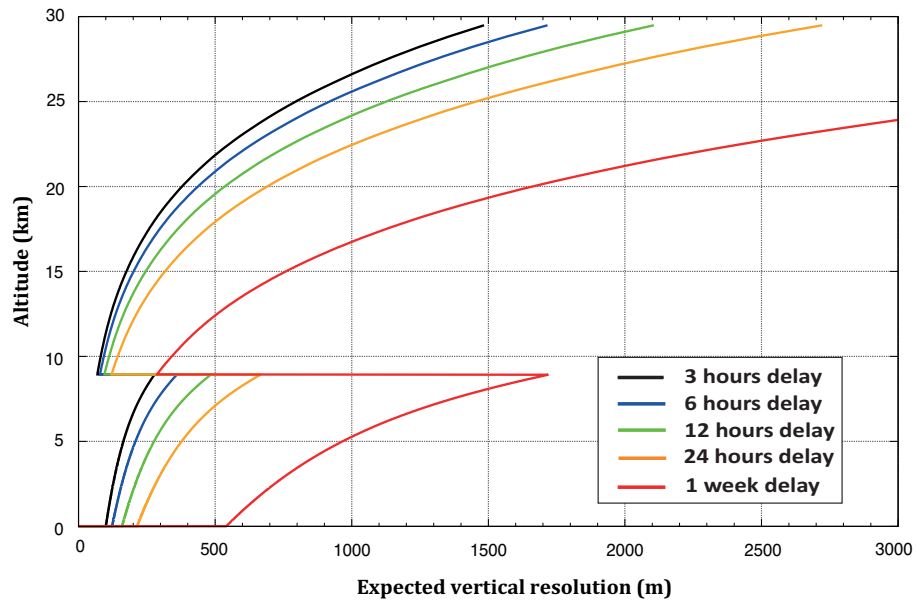
Figure 3. Impact of the time delay between landing and analysis on the expected vertical resolution of AirCore-HR, for a storage time of 3 hours (black), 6 hours (blue) and 12 hours (green), 24 hours (orange) and one week (red).
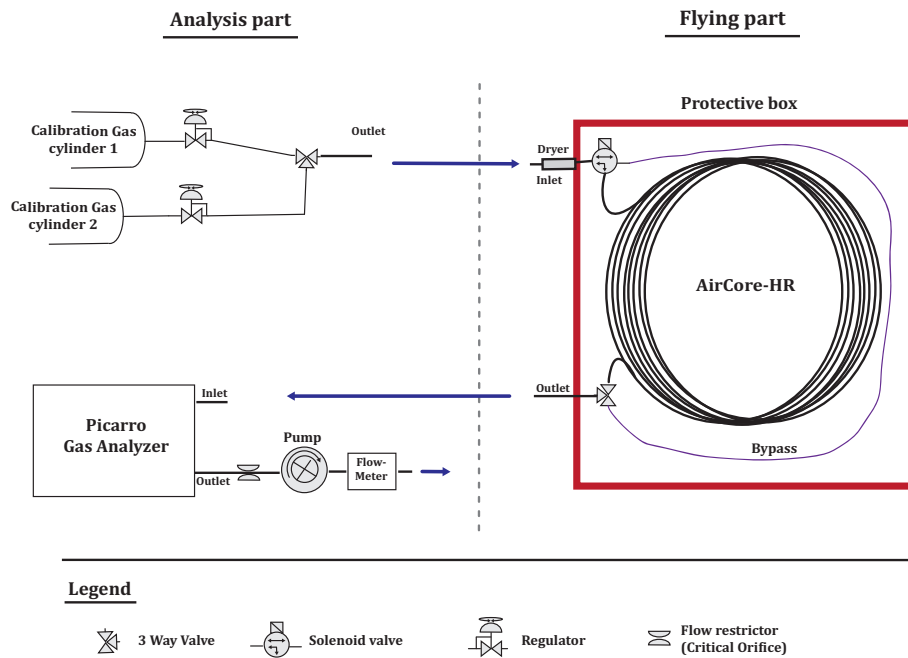
Figure 4. Overview of the AirCore-HR and analysis system
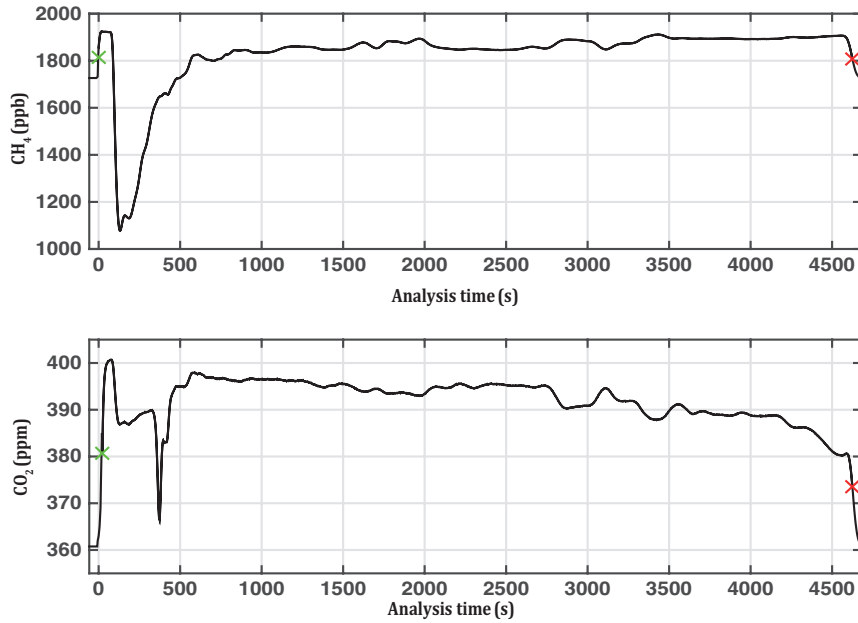
Figure 5. Picarro analysis of the AirCore-HR sample from "EdS-Stratéole" flight on 29 August 2014. (top) CH_4 mixing ratios as a function. of the analysis time in seconds, (bottom) CO_2 mixing ratios as a function of the analysis time in seconds. The selected starting point of the sample is marked with a green cross and marks the 0 of the analysis time; the selected ending point of the AirCore-HR sample is marked with a red cross.
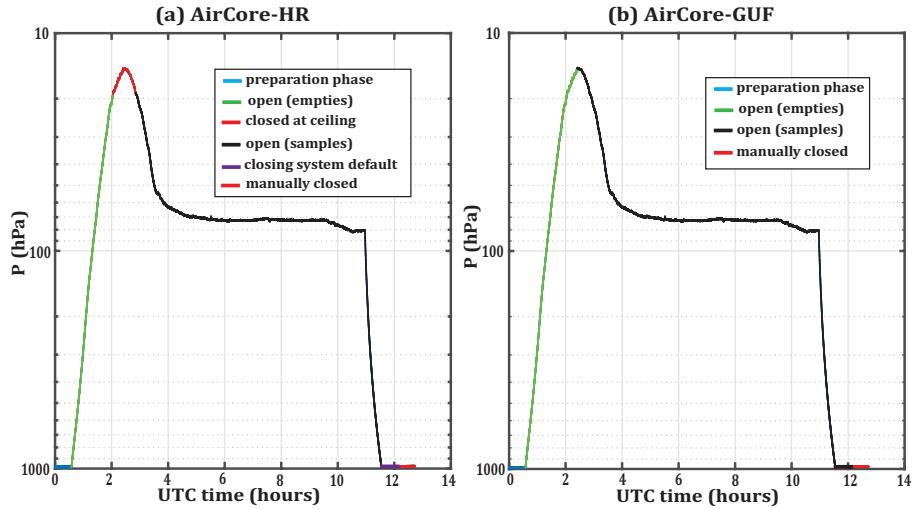
Figure 6. Flight plan from the StratoScience 2014 "EdS-Stratéole" flight on 29 August 2014 with main operating states of (a) AirCore-HR, (b) AirCore-GUF : the preparation phase, on the ground before flight (blue), ascent phase (green), descent phase (black), closed (red).
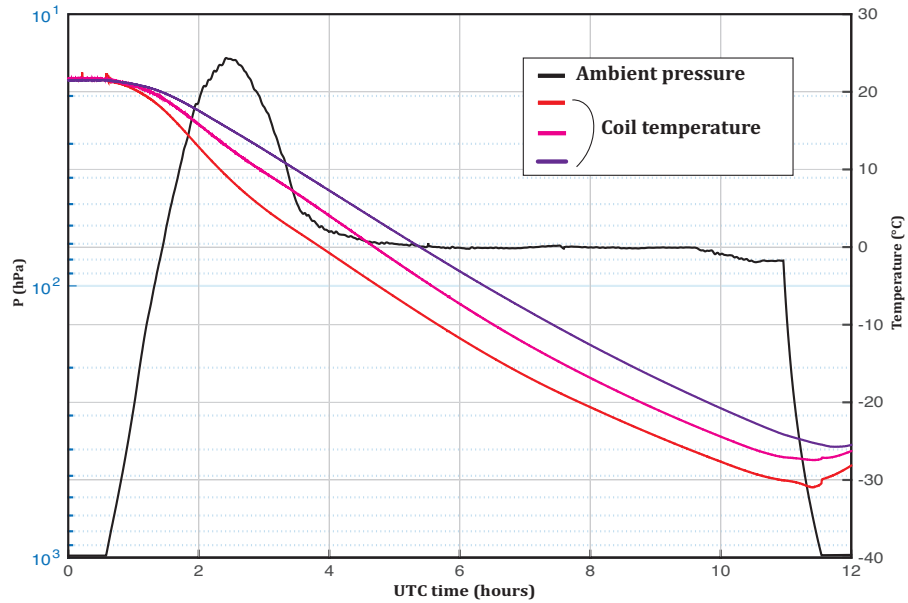
Figure 7. Recorded temperature from the 3 probes on the AirCore-HR and ambient pressure during flight "EdS-Stratéole" flight on 29 August 2014. The 3 temperature probes (red, pink and purple lines) are presented in °C in function of UTC time, the temperature axis is located on the right side. Ambient pressure (black line) is presented in hPa in function of UTC time, with the vertical scale on the left side.

Table 1. Characteristics of the AirCore-HR.

Tube	1/4 in. tube	1/8 in. tube
Diameter (inches/cm)	0.25 / 0.635	0.125 / 0.3175
Width (inches/cm)	0.02 / 0.0508	0.02 / 0.0508
Inner diameter (inches/cm)	0.21 / 0.5334	0.085 / 02159
Length(m)	100	200
Volume (cm^3)	2235	732
Weight (kg)	7.48	6.82
Overall payload		
Overall payload Volume(cm^3)	2967	
Overall payload Weight	14.30	
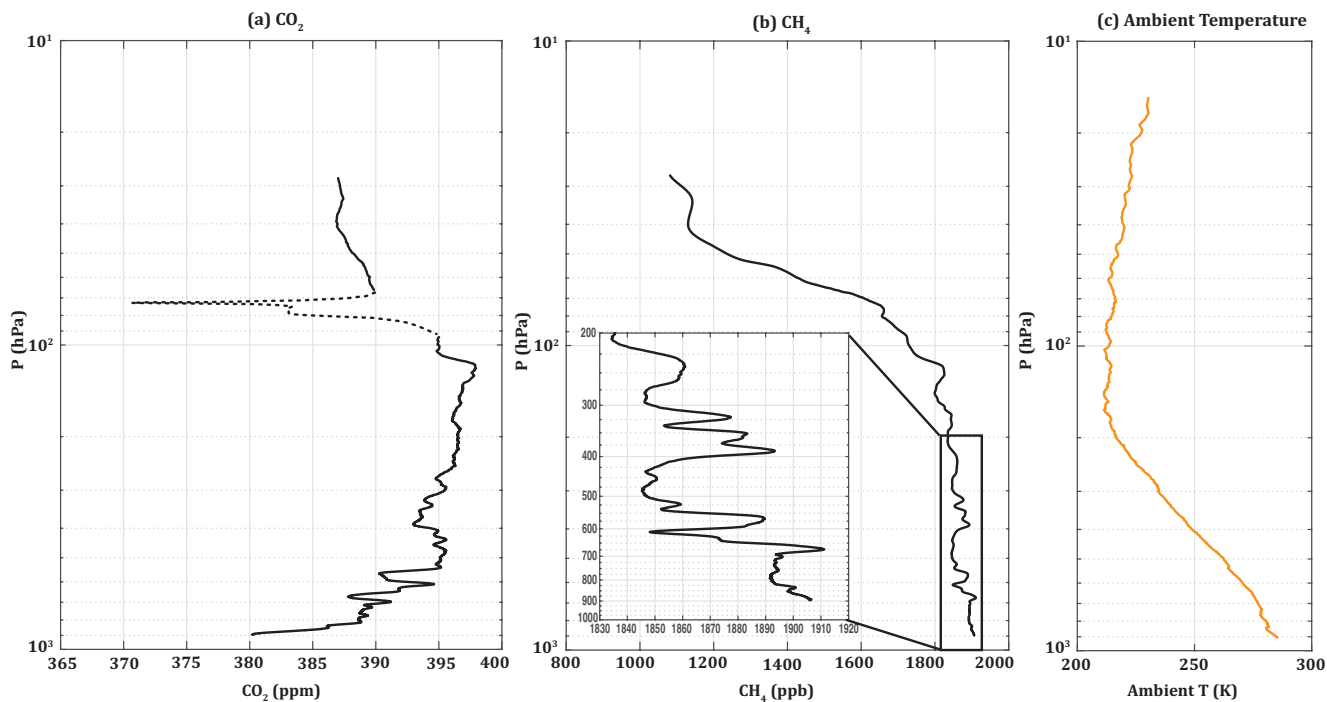
Figure 8. Vertical profiles retrieved from the air sampled with AirCore-HR on the "EdS-Stratéole" flight on 29 August 2014. (a) CO_2 (ppm), (b) CH_4 (ppb), (c) ambient temperature (K). The dotted line in the CO_2 profile corresponds to a part of the profile with unrealistic CO_2 values due to flight trajectory (see text)

Table 2. Values of the calibrated gas standards using NOAA's "WMO Scale" reference. The air of the two reference tanks used in this study was measured at LSCE with a Picarro G2401 calibrated with a scale of 6 tanks from NOAA/ESRL. The table shows the reproducibility of the measurements and standard deviation over 3 measurements made on a 15 day period.

Low Concentration standard:
CO_2 360.85 ppm $\pm$ 0.008 ppm
CH_4 1726.95 ppb $\pm$ 0.163 ppb
High concentration standard:
CO_2 401.31 ppm $\pm$ 0.004 ppm
CH_4 1922.33 ppb $\pm$ 0.168 ppb
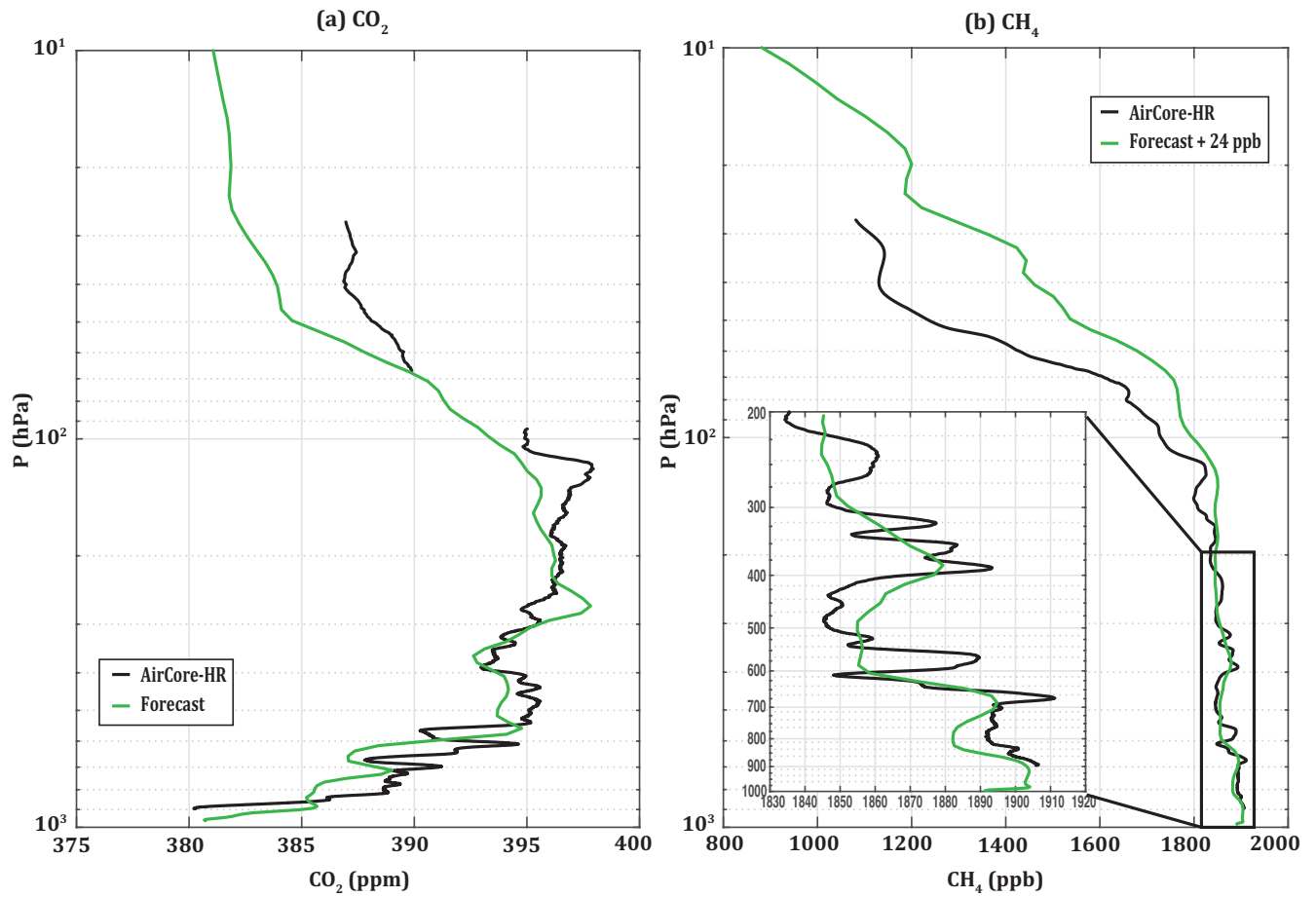
Figure 9. Comparison of AirCore-HR (a) CO₂ and (b) CH₄ vertical profiles (black) with co-located high resolution forecast (green) from CAMS-ECMWF at landing coordinates on 29 August 2014 at 12 UTC. 24 ppb were added to the CH₄ high resolution forecast.
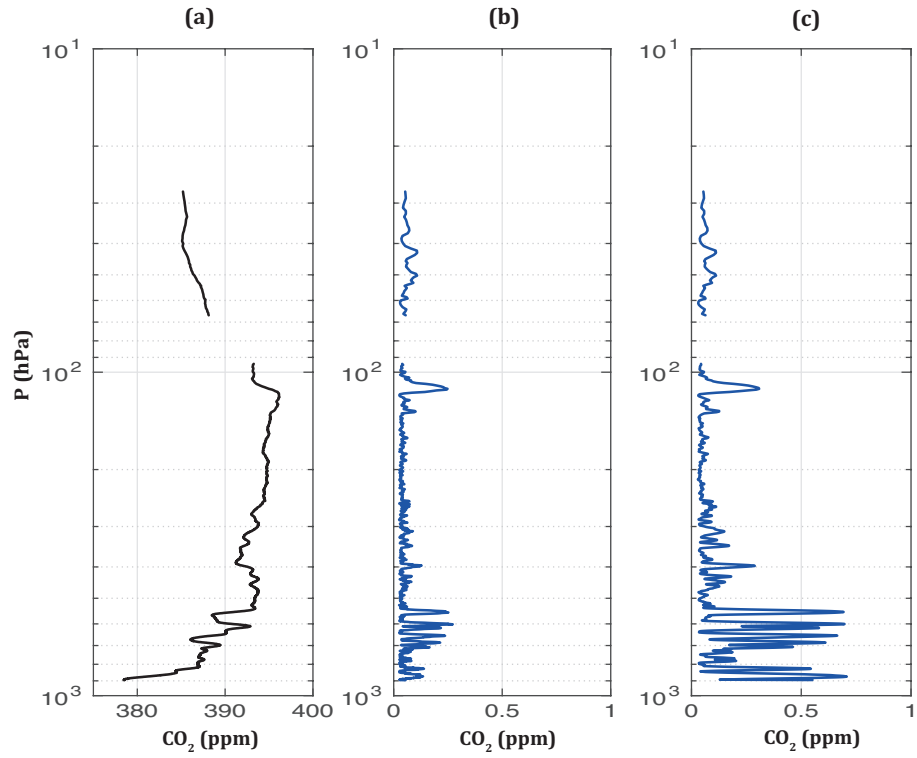
Figure 10. (a) AirCore-HR CO_2 vertical profile, (b) CO_2 uncertainty without taking the potential loss of air into account (see text uncertainty (v)), (c) overall CO_2 uncertainty.
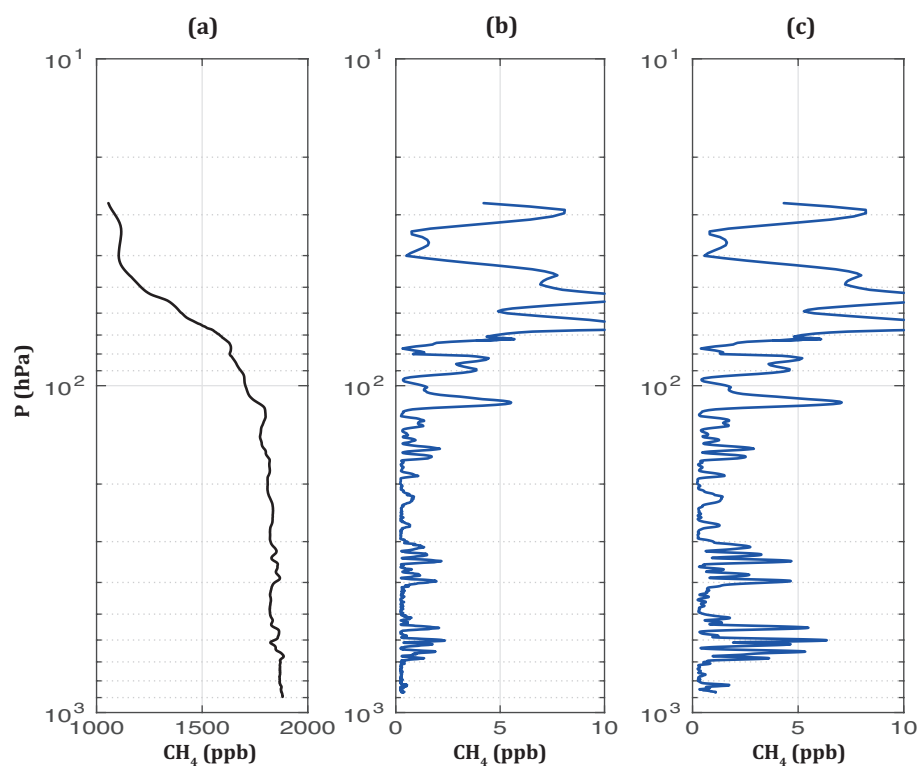
Figure 11. (a) AirCore-HR CH$_4$ vertical profile, (b) CH$_4$ uncertainty without taking the potential loss of air into account (see text uncertainty (v)), (c) overall CH$_4$ uncertainty.
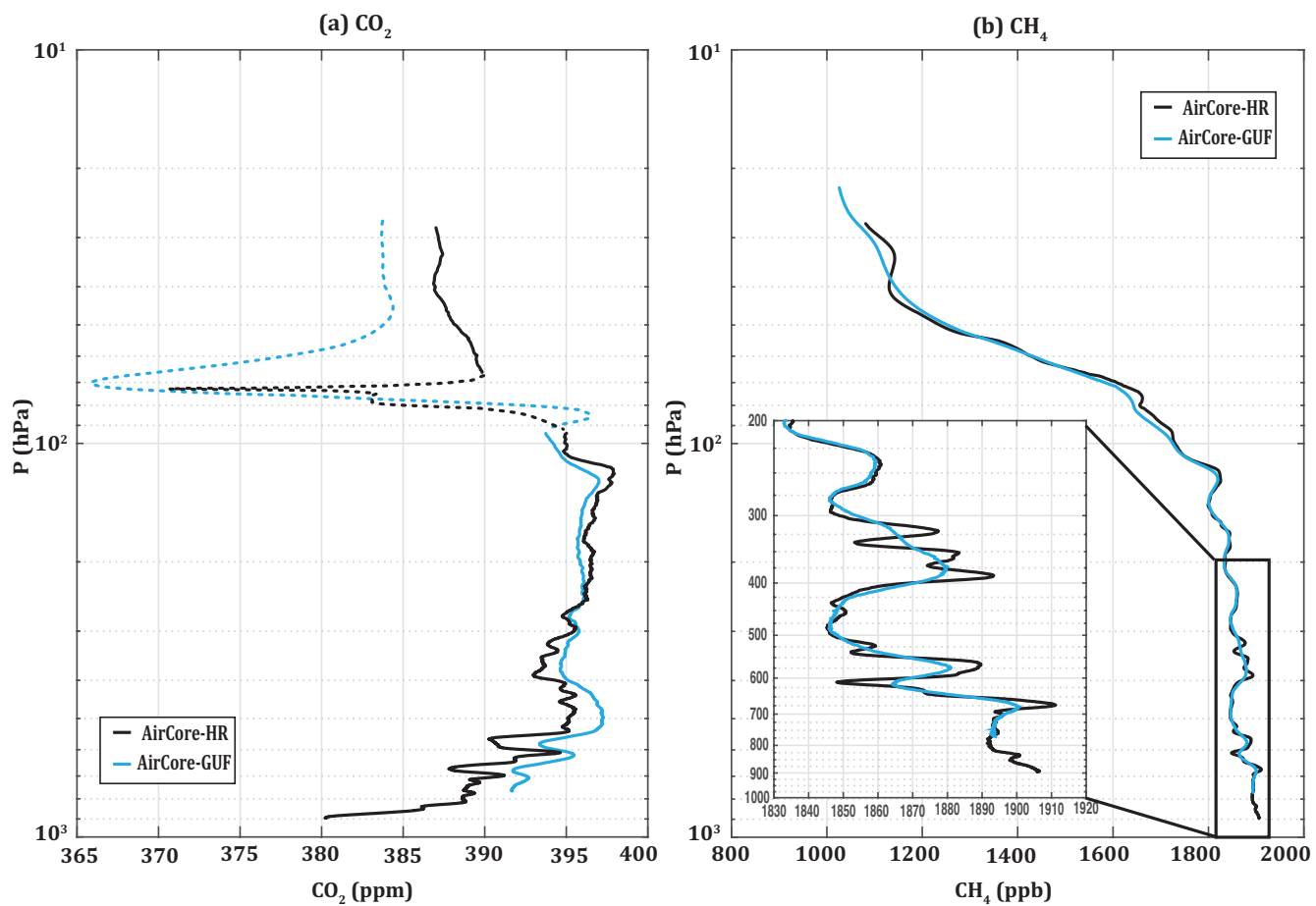
Figure 12. Vertical profiles retrieved from the air sampled with the AirCore-HR and an AirCore-GUF on the "EdS-Stratéole" flight on 29 August 2014. (a) CO_2 (ppm), (b) CH_4 (ppb). The dotted line in the CO_2 profiles correspond unrealistic CO_2 values that where sampled during the long plateau phase (see text).
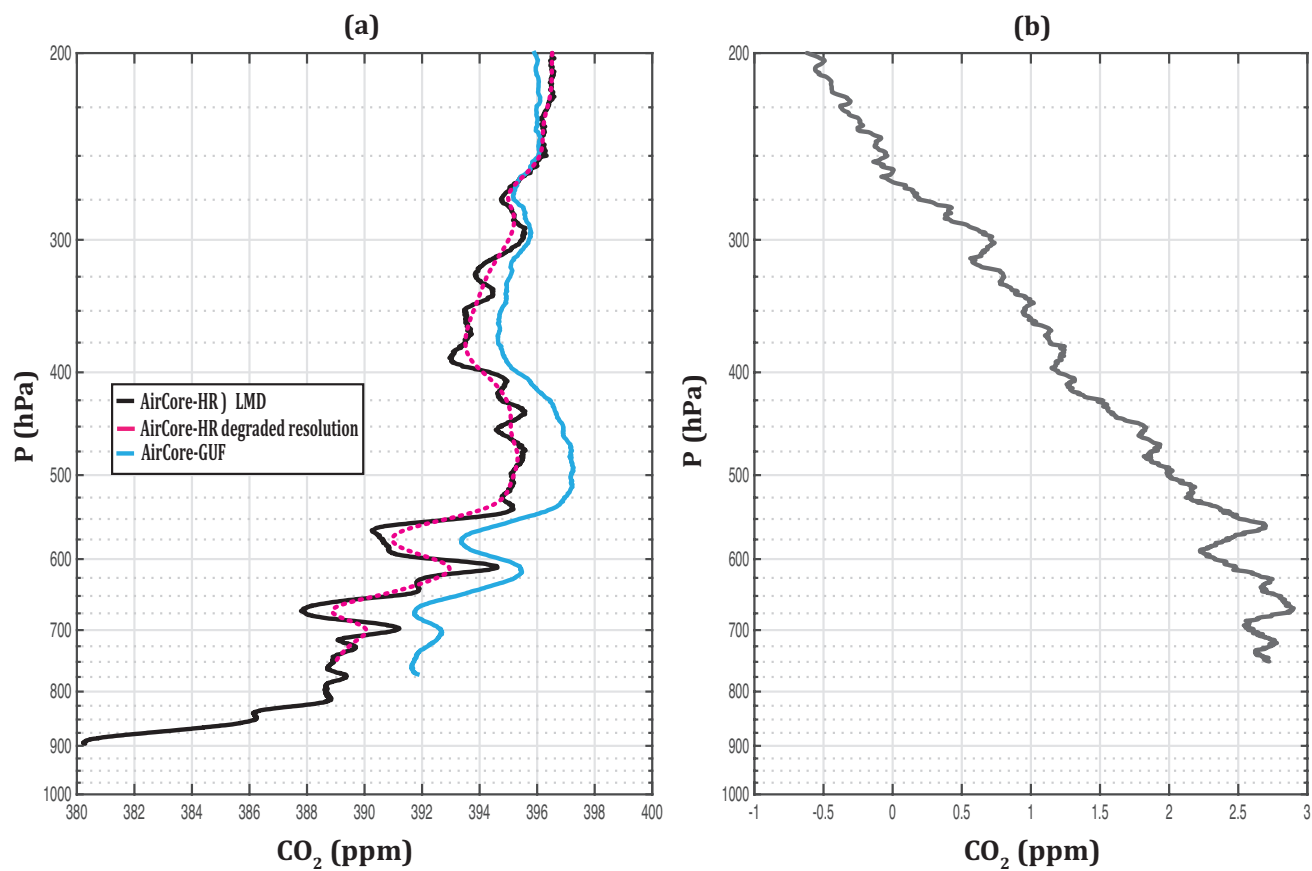
Figure 13. (a) CO_2 vertical profiles from AirCore-HR in full resolution (black), AirCore-HR "degraded resolution" (pink), AirCore-GUF (blue). (b) Residual of the difference AirCore-GUF - AirCore-HR "degraded resolution".
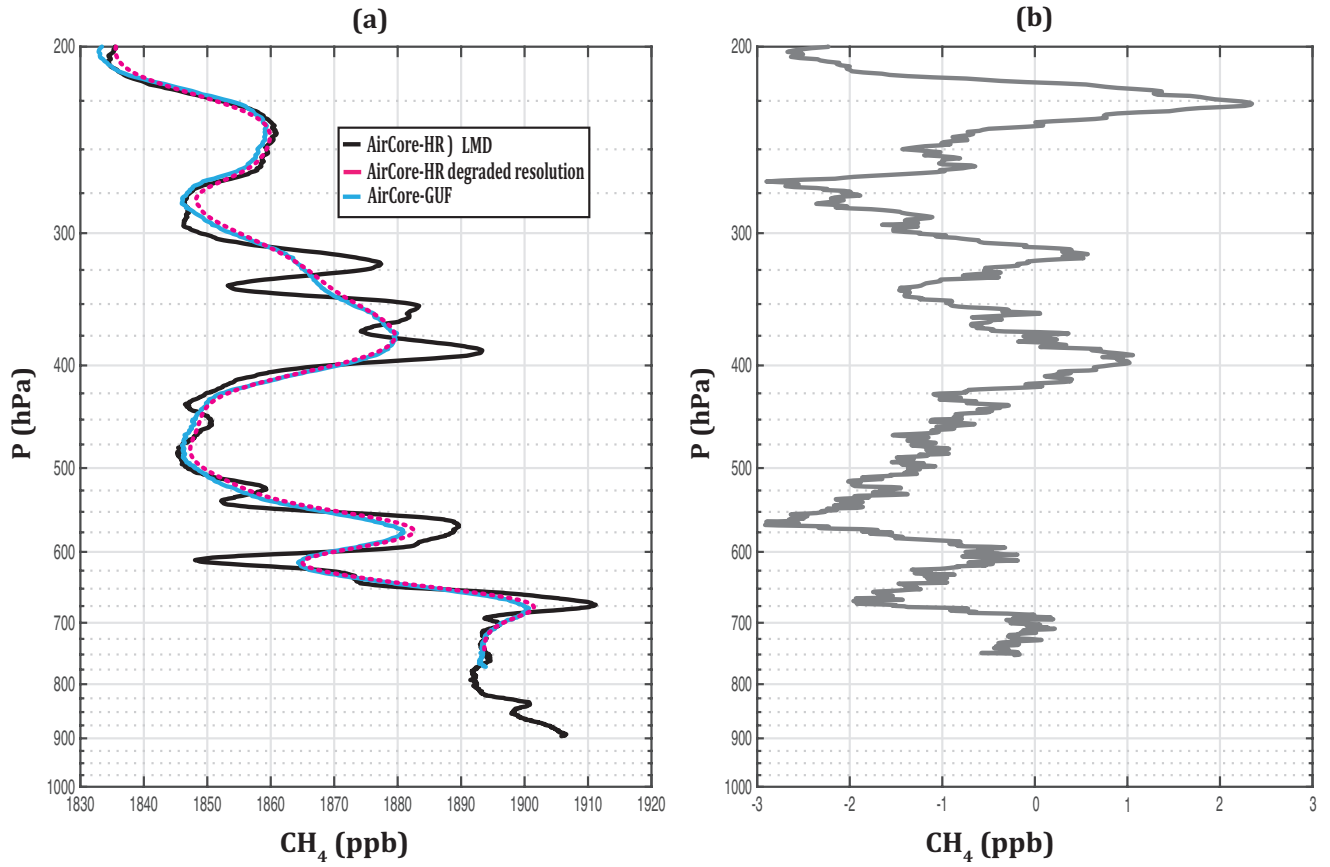
Figure 14. (a) CH₄ vertical profiles from AirCore-HR in full resolution (black), AirCore-HR "degraded resolution" (pink), AirCore-GUF (blue). (b) Residual of the difference AirCore-GUF - AirCore-HR "degraded resolution".