Authors' response to reviews of Local and Regional Scale Measurements of CH_4, $\delta^{13}CH_4$, and C_2H_6 in the Uintah Basin Using a Mobile Stable Isotope Analyzer

Prepared on 2015 09 07

Summary by Rella et al. to comments: The two referees and the editor have provided a great deal of useful and constructive criticism, which have led to many improvements and clarifications in the manuscript. In nearly all instances the corrections were incorporated into the manuscript. We believe we have addressed all the concerns raised by the referees and editor.

Anonymous Referee #1

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General.

This paper is a detailed account of the function and testing of a CRDS instrument for mobile field measurement of d13C in CH4. Rapid isotopic analysis, using a vehicle mounted analyser deployed in the field, of methane emissions from both anthropogenic and natural sources is an extremely valuable technique: this paper thus represents a major advance in our ability rapidly to identify sources of methane emissions in the field. The work is well detailed and well argued, and fully deserves to be published.

Specific Comments.

General comment. The paper is presented by the instrument manufacturer. Though there could be conflict of interest, the origin of the work is clearly stated, the study is factual and carefully detailed, and the paper thus stands solely on its scientific merits.

L25. Trivial but annoying – Uinta / Unitah basin? Many times in the paper. . ...Choose one spelling? – or like Lawrence of Arabia, when confronted on this by his publisher, add many more spellings and make it interesting. You in Terra?

[Rella et al.] Good catch. We have elected to follow Karion et al. (2013) with the spelling Uintah. Corrected in the text.

L39 – GWP 28-86x – explain this more fully: it means over differing timescales but as written here looks like error. The Howarth paper is probably not the best to cite on the fraction equivalence. Also maybe mention in more detail that gas displaces coal, but gas leaks at Unita percentages make the greenhouse benefit very dubious.

[Rella et al.] Excellent comment.

- Rewrote the passage to clarify the effects of methane on 20 and 100 year time scales
- Removed Howarth reference.
- Highlighted comparison to coal

L74 – maybe cite Dlugokencky et al 2011 here?

[Rella et al.] Excellent. Reference added.

L81 – maybe cite Lowry et al. 2001 on early use of d13C to constrain London methane emissions

[Rella et al.] Another excellent reference. Reference added.

L90 – use of isotopes: should probably cite Fisher et al., 2006 and 2011 here – GC-CF-IRMS technique is the clear option compared to CRDS and needs comparision somewhere in the paper. Both methods have their uses – CRDS gives moderate precision but is rapid and field-deployable, while GC-CF-IRMS involves the limitations of bag sampling for offline analysis but has 0.05 per mil precision. Looking forwards, it is likely that both techniques would have their future place in a good field campaign.

[Rella et al.] We agree completely that the methods are complementary. The two references have been added, as well as a summary of GC-CF-IRMS performance as achieved in the Fisher 2006 reference.

L100 – probably it would help to add two or three lines as a brief overall explanation of CRDS, for those new to the technique.

[Rella et al.] Excellent suggestion. A short description has been added, to complement the longer description L156-L183.

L112– give metric equivalents, as many readers might actually be from outside the USA or the gas industry.

[Rella et al.] The definition of units and conversion to metric has been provided in the revised manuscript.

L272 – 1% 'more than sufficient' – can that be substantiated by a few lines in the supplementary material?

[Rella et al.] The following sentence was added to the text of the manuscript: While this degree of accuracy (20 ppb at 2,000 ppb of CH4) is insufficient for global methane measurements, it is adequate for the purposes of this study in which the typical methane enhancement was higher than 1,000 ppb.

L289-313 and also L369-382 – I have not had time to work though this algebra carefully. It looks OK, but my experience is that triple checking subscripts is useful.

[Rella et al.] This is good advice. We have rechecked the algebra.

L314 – model, vendor??

[Rella et al.] Vendor information added.

L432 – high degree of stability – could we have some numbers on this? – long periods of time? i.e. days? months? In constant temperatures or in large diurnal variations in field deployments when the instrument is in the sun and bumped up and down?

[Rella et al.] We have clarified this statement as follows in the manuscript:

Given this degree of stability, and if care is taken to ensure a somewhat stable environment, we expect that with a properly-crafted calibration scheme this instrument can be used for long-term (months or years) in situ atmospheric monitoring of $\delta^{13}CH_4$.

L450 – Oxygen and Argon: yes, but natural gas wells can leak helium and argon – could get unexpected local effects close to wells before mixing takes place? maybe it's too small to be significant?

[Rella et al.] This is an important clarification. The following text was added: We note that while it is possible for Argon or other inert gases (e.g., Helium) to be present in natural gas plumes, we expect that the low concentrations that would be present in the dilute downwind plume to affect the isotope ratio measurement negligibly.

L474– a little more detail on CO?

[Rella et al.]. We have added the following additional information to the section on CO: The spectroscopy of this simple molecule is extremely well understood. Our spectroscopic modeling shows that at the same concentration as methane, the effect on the measured isotope ratio is expected to be negligible. The effect could be more pronounced in an atmosphere rich in carbon monoxide.

L497 – driving speeds? In Utah maybe 90mph, but in Europe can you do 10kph and traverse plumes slowly or even stop and creep back and forth through them? The custom flow system is excellent and innovative – may prove to be very useful in rapid field mapping of plumes of all origins. This is a major merit of the paper.

[Rella et al.] At slower driving speeds, the meander of the plume across your sampling point leads to rapid variations in the methane signature, so simply driving slower is rarely effective at improving the isotope analysis. We have generally found it to be effective to traverse the plume cleanly so that samples of the plume as well as the background air are obtained in a single long sample that can then be analyzed. We added a typical range of driving speeds (5 – 30 m / s) to the text.

L528 – here or later in Fig 16 maybe a separate paragraph in discussion to introduce Keeling and Miller/Tans plots and perhaps cite Pataki et al, 2003 as well as Miller and Tans?

[Rella et al.] This is an excellent suggestion. The following modification has been made to the text:

To quantify the isotopic signature of each individual plume, we have followed the analysis described by Miller and Tans (2003), which is analogous to the Keeling analysis performed in

Pataki et al. (2003). Both analysis methods rely on a simple two-member mixing model for source and background gas…

L581-603 – again, haven't checked the subscripts line by line. . .looks OK but..

[Rella et al.] Equations checked again

L611 – modified Air Core system - very nice. . .

L679 – maybe some more detail here?

[Rella et al.]: the following was added to this paragraph: At our typical driving speeds of 15 m / s, 15 seconds corresponds to 225 m of distance; thus, our algorithm defines a 'local' source as producing a plume narrower than 225 meters.

L700-710 – valuable conclusions that demonstrate the power of the technique.

L739 – update needed???

[Rella et al.]: The text has been updated with the February 2013 value, and figure 16 was updated. The Monte Carlo analysis was rerun using the 2013 values for isotope ratio and concentration, and the resulting outputs of the Monte Carlo changed by at most 1-2%. The values in the text represent the updated numbers.

Figures are generally clear, if pedestrian in presentation. In Fig 15 the scale needs units. Overall, the figures could be brushed up a little.

[Rella et al.] Units have been added to figure 15. The figures will undergo a final review and polish prior to final submission.

Conclusion

This is an important presentation of the use of what will likely become a widely used instrument in the field. The implications both for atmospheric science and the natural gas industry are significant, and should lead to identification and thus to reduction of emissions. The paper should be published with minor revisions.

Anonymous Referee #2

Received and published: 16 June 2015

This is an interesting paper that is certainly worth publishing as it describes in detail the development of a new method to measure ethane concentration in the atmosphere, as well as expanding on the measurement of methane from a mobile platform. The abstract and introduction are generally concise and describe the paper well. The flow of the paper is good. The figures are also generally good; I have some minor changes suggested below. Overall, I feel that this paper requires minor revision. Following are a number of queries and suggested changes.

1. In the introduction, no mention is made of what background CH4 and C2H6 levels are in the US. This would be good information, as well as giving the range of values for both as observed in the Uintah. For example, background C2H6 is ∼0.5 ppb. The stated measurement precision of the analyzer is given +- 20 ppb for C2H6: that doesn't sound so good. From figure 16b it appears that C2H6 levels around gas wells may be ∼1 ppm. I think that some discussion of background vs. observed concentrations of both gasses would be interesting in the Introduction.

[Rella et al.] This is a very interesting and useful comment. The following text has been added to the introduction.

Typical concentrations of methane and ethane in the clean continental atmosphere is 1.7 – 1.9 ppm and 0.5 – 2.0 ppb, respectively, but in regions where emissions of these gases are high, concentrations can rise as high as ~10 ppm CH4 and ~1,000 ppb C2H6. At these levels, this instrument is useful for individual source characterization as well as quantification of the overall atmospheric signature in a given region, activities that are crucial to regional source apportionment.

2. A lot of effort is put into justifying the need for the "long" tube in the search for the regional signature (as opposed to the "short" tube used for in-field data playback), i.e. that the results capture nearly all of the data seen in the field. While this is interesting and presents an intriguing way to capture data without the need to have an analyzer present, no evidence is shown that supports that the result is "better" than the results for the short tube. There are statements that the

advantage is that standards can be used more frequently during the long tube playback, but that that was not done here, etc. It would be interesting to see how the results compare with what was measured in closer to "real time".

[Rella et al.] The referee brings up an interesting question. It is important to remember that the 'short' tube represents the analysis of individual plumes, not of well-mixed regional background air (more on that below, in #3). However, we could compare the well-mixed regional background air as measured on the vehicle to the measurement made from the long tube. In that instance, the increase in the measurement time is about a factor of 10 (from 2 hours to 24 hours), leading to an improvement in precision of $\sqrt{10} \sim 3$X. In addition, because no standards were available on the vehicle, we were unable to calibrate the instrument.

The following text has been added to manuscript: We estimate that by using this larger AirCore we have improved the precision of the regional air measurement by about a factor of 3 relative to the in-vehicle measurement, without the need to carry a compressed air cylinder in the vehicle.

3. In figure 12 the δ13CH4 values for the Uintah gas and oil that are used as "typical" values are based on the centroids/averages of the 28 tube short samples. It seems to me that no matter how well each sample characterizes a single source, the samples are still going to be made up of mixtures of at least "regional air", "gas air" and "oil air". It appears likely that the actual isotope ratio for oil air will be lower than the average, and similarly the gas air will be higher than the average. Similar arguments could be made about the C1/C2 ratio. This should be stated. It is interesting to look at the Keeling plot in regards to this, as it appears to be showing this, albeit the statistics on the fit will not be good. You state that there is no fractionation (p. 4882, 1st paragraph), so the difference may be small.

[Rella et al.] This is a very important point. The key point is that the two-member mixing model (of which Keeling and Miller-Tans analyses are subsets) gracefully handles this effect by taking out the effect of the background. This point has been made explicitly in the text by adding the following to the discussion of Miller Tans analysis :

Note that the background gas in this analysis (and contained in the AirCore tubing) is the regional air sample at the locale where the plume was measured encountered immediately before and after the plume was encountered. As such, it contains not just continental background air, but also air contaminated by other sources in the region. The advantage of the Miller-Tans analysis for the isotopic signature (and the slope analysis for the ethane to methane ratio) is that variations in this background from site to site do not affect the derived source signature for the plume.

4. I think that the "regional atmospheric signature" section needs to be reorganized. It starts with extensive equation development, breaks suddenly (to me) into a description of data collection (page 4888, line 10) and then breaks back into more equation development and results after that (p. 4889, line 18). This was confusing and should be reorganized.

[Rella et al.] We understand and appreciate the assessment of the referee on this section. We carefully considered other organizations for this section, but in the end, we decided that the existing organization [that is, 1) a theoretical treatment to link regional emissions ratios to observable tracer ratios and quantities, 2) an experimental section to describe how those tracer ratios can be quantified from the vehicle, followed by 3) a description of the analysis algorithms used to process the data was clearest to the reader. Introducing #3 prior to #2 would be out of context for the reader, and introducing #2 prior to #1 would again leave the reader without the appropriate context to understand this section. We respectfully disagree with the referee about the suggested reorganization, and have left the order largely intact.

5. Minor: There are a few places where the authors state things like: " for this experiment, we will assume that the term C'(t) is zero" (p. 4876, line 2). Obviously the assumption is that the contribution from this term is negligible, but this should be stated (and a reason given?).

[Rella et al.] Excellent observation. The actual intent of this section was to demonstrate the performance of a specific calibration scheme rather than make an assumption about the size of C'. The text has been clarified as follows:

In this section we describe an experiment in which we track the drift of the instruments. In this experiment, we will ignore the contribution of the term C'(t). The two remaining drift terms c_0 (t) and χ(t) have a markedly different dependence on concentration. Therefore, to effectively track drift, we will use two bottles with different CH4 concentrations. Note that a more accurate calibration scheme would be to track the term C'(t) by employing a third bottle with a substantially different isotope ratio than at least one of the first two bottles. Although the algebra becomes more complicated, the outcome is similar to calibration using two bottles

6. Minor: My understanding is that Air Core is a) patented and b) actually a tube closed at one end. Wouldn't it be more correct to describe the two tube-based systems shown here as being "based on Air Core"?

[Rella et al.] Very good point. The text was edited in the following way:

This technique is based on a technology called AirCore that was first developed by NOAA as a simple and effective way to measure the vertical profile of trace gases in the atmosphere (Karion et al. 2010; and Tans 2009).

7. Minor: Equation 1 looks like the Allan variance, not the Allan standard deviation.

[Rella et al.] Correct. The text was modified to clarify this point.

8. Minor: In page 4872, line 23 mass flow controller should have (MFC) after it to clarify what MFC is in Figure 4.

[Rella et al.] Corrected in the manuscript.

9. Minor: The equation numbering is wrong in the Supplement. There are two Eq. 2's and 3's. And S Eq. 5 mentioned in the main body does not match the supplement equation

[Rella et al.] Corrected in the manuscript.

5. Even more minor: the equations in the supplement are numbered with SM, while in the supplement they start with an S.

[Rella et al.] Corrected in the manuscript.

10. Minor: on page 4872, lines 7 to 8 – it appears that the authors wanted to supply model and vendor information.

[Rella et al.] Corrected in the manuscript.

11. Minor: p. 4878, line 3 – δ13CH4 is not a mole fraction.

[Rella et al.] Corrected in the manuscript.

12. Minor: p. 4886, line 22, it is stated that D=0.2 cm2/s, without attribution. This information is given in the supplement, but not here.

[Rella et al.] Reference added to the manuscript.

M. Hamilton (Editor)

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This is an interesting paper, that reports important and topical work. However I wish to make some criticisms of an editorial nature in order to expedite the process of revision for consideration for publication in AMT. The paper has many colloquial usages of English which need to be made more formal, and in addition, the fact that the authors are from the company which made the instrument shows through - and I think detracts from the value of the paper. My specific comments are below and the line numbers pertain to the original version of the manuscript which I believe the authors have. These comments may be made redundant by the response(s) to the referees, but should be borne in mind when making those responses.

[Rella et al.] We value these constructive criticisms from the editor. While we strove to remove jargon, colloquialisms, and biased language from the manuscript, this language crept into certain passages. We are grateful to the editor for catching this on many occasions; we have again reviewed the manuscript with this criticism in mind to further improve the language.

General comments

a) The words innovative and novel are overused. This should be reduced to just two (at most) usages; I'd suggest once in the abstract and once in the conclusion - and no more.

[Rella et al.] Agreed. The manuscript has been edited to reflect this recommendation.

b) Section 2 should be broken up with the CRDS in one section (#2) and the results from the Uintah basin in another (#3). This would be more consistent with the title of the paper.

[Rella et al.] This was the original intent – this division was lost at some point in the manuscript preparation. Corrected in the manuscript.

Abstract

L19; the words "highly precise" are redundant

[Rella et al.] Phrase removed.

L23,24; "... and a novel onboard gas storage and playback system." This text seems to be repeated in the second to last sentence of the abstract. I don't think this is the case, but it should be made clear that there are two playback systems.

[Rella et al.] The second-to-last sentence in the abstract has been modified as follows:

Furthermore, regional measurements of the atmospheric CH4, d13CH4, and C2H6 signatures throughout the basin have been made, using continuous sampling into a 450 meter long tube and laboratory reanalysis with the CRDS instrument.

L29; the concept of a "signal present in the ground" is too vague.

[Rella et al.] Replaced this with 'signatures of the gases found in the wells.'

Main body of manuscript

L36,37 grammatical problem (production ... WAS ... viable)

[Rella et al.] Corrected.

L37 the word "there" is missing

[Rella et al.] Corrected.

L63 "these OTHER emission" ???

[Rella et al.] Text clarified as follows:

While the Uintah basin is fairly simple from the standpoint of methane emissions, with a small population (~60,000) and no significant sources of methane apart from oil and gas extraction, the oil and gas producing area in the Denver-Julesburg basin is largely co-located with other sources of methane, such as landfills and concentrated animal feeding operations.

L74,75; grammatical singular/plural problem

[Rella et al.] Corrected.

L82; do you mean "firn" or "fern" ? I think the former!

[Rella et al.] Good catch! Corrected.

L133; what are "end member populations"?

[Rella et al.] Replaced with 'individual source signatures'

L142; by THIS instrument

[Rella et al.] Corrected.

Section 2.1; You seem not to make the connection between ringdown time and molecular absorbance.

[Rella et al.] This is a critical omission! The text has been modified as follows:

CRDS is a method in which laser light is coupled into a resonant optical cell. The decay rate of the optical power in the cavity is a direct measurement of the total loss, which includes both absorption loss due to the gas mixture contained in the optical cell and the loss of the mirrors in the system. Two separate lasers…

The instrument employs precise monitoring and control of the optical wavelength, which delivers sub-picometer wavelength targeting on a microsecond timescale. When the light resonates in the optical cell, the laser is turned off. The resulting decay of optical power, called a ring down, is measured with a fast photodetector. From the ring down decay time the total absorbance of the system is derived from the equation $\alpha = 1/c\tau$, where α is the absorbance, τ is the ringdown time, and c is the speed of light. Ring down events are collected…

L174-176; It is unclear as to whether the species CH4, CO2 & H2O are analytes or "other key atmospheric constituents"

[Rella et al.]. They are both! But the text should be clearer. We have edited it as follows:

The right panel of Fig. 1 displays the spectra of key gas species, which includes the five analytical species and other atmospheric constituents that absorb in the 6057 wavenumber region…

L184; grammar - "are" not "is"

[Rella et al.] Corrected.

L188 - 191; There are words missing from this sentence

[Rella et al.] Corrected.

L200; is the word "spectra" missing in the first sentence?

[Rella et al.] Corrected.

L200; delete "experimental" - it’s redundant

[Rella et al.] Corrected as follows: Using a prototype instrument, high resolution spectra over a range…

L203; the word "only" is repeated unnecessarily

[Rella et al.] Corrected.

L205; it’s puzzling as to why you need to refer to the HITRAN version in 2010

[Rella et al.] Parenthetic comment removed.

L207; grammatical singular/plural problem

[Rella et al.] Corrected.

L211,212; rephrase this to be less colloquial

[Rella et al.] Corrected as follows:

It is possible that the removal of the 13CH4 spectrum is imperfect, leading to a cross-interference between 12CH4 and 13CH4. This cross-interference can be dealt with in a straightforward manner during the calibration of δ13CH4, as discussed below.

L216; in what sense does CO2 have amplitude?

[Rella et al.] Replaced amplitude with ‘strength,’ a standard spectroscopic quantity,

L220; "purpose OF"??

[Rella et al.] Corrected.

L232; what is a "stronger dynamic range"?

[Rella et al.] Corrected to:

This line affords the instrument a much larger dynamic range…

L239; You measure something but not the bottle! (Similarly L272 - what is the "accuracy of a tank"?)

[Rella et al.] Corrected as follows:

We have performed a basic assessment of the instrument by measuring gas from a high pressure cylinder containing 1.78 ppm CH4 for 18 hours.

And

The analytical accuracy of the gas mixture in this tank is 1%.

L246,248; This seems to be unnecessarily repetitive (see §2.1)

[Rella et al.] Good observation. The last sentence has been removed.

L263; linear relationship with what?

[Rella et al.] Corrected as follows:

The peak height α does not always vary linearly with gas concentration, but in most situations in near-infrared resolved ro-vibrational optical spectroscopy, the relationship is substantially linear:

L270; the footnote doesn't really explain ppb/cm as a unit of loss - is something that can be better supported by a suitable reference?

[Rella et al.] We don't believe a reference will clarify this definition. Instead, to clarify, the footnote language has been edited:

ppb/cm, or parts-per-billion per cm, is a unit that describes optical absorbance as a fractional loss of optical power per unit distance.

L285; why mention just water here - wouldn't this apply also to dilution by the likes of O2 or Ar?

[Rella et al.] Excellent observation. Corrected as follows:

2) Dilution by other gas species, such as water vapor, oxygen, or argon, occurs to both species equally (as a % of each species' dry mole fraction), which means that δ13CH4 is unaffected by dilution.

L288; the expression "net lineshape effect between the two species" is too vague

[Rella et al.] Clarified as follows:

3) Spectral line shape effects due to other gas species are likely to have similar effects on the two methane species. δ13CH4 is affected only by differences in the line shape effects between the two species.

L296; please decide if this is to be an in-line equation or not

[Rella et al.] Corrected.

L309; Is this not a peak height ratio rather than a loss ratio?

[Rella et al.] Corrected.

L329,330; it is unclear which are the standard calibration constants give the specific symbols.

[Rella et al.] Corrected.

L339; " four standards" should be " four isotop ratio standards"

[Rella et al.] Corrected.

L360,361; What is a drift in the wavelength axis?

[Rella et al.] Corrected as follows: Drift in the laser wavelength reported by the wavelength monitor also tends to cause errors in the measured peak height that is proportional to the peak height,…

L367; similarly, what is nonlinearity in the absorbance axis?

[Rella et al.] This is described in the supplement:
(i.e., nonlinearity in the absorbance axis and cross-talk between 12CH4 and 13CH4, as described in the supplemental material)

L376; c_0(t) should be defined here, and not just in the supplementary material.

[Rella et al.] Corrected.

L377 - 379; this should be turned into proper text.

[Rella et al.] Corrected.

L386,387; missing word in this sentence

[Rella et al.] Corrected.

L399; this paragraph confuses the reader as to whether this is two or three bottle testing

[Rella et al.] Corrected as follows:

There are two bottles in the experiment we performed, shown in Fig. 6: a high…

L429; perhaps a sentence here, cross referenced to §2.3, which clarifies the relationship between the two Allan variation graphs presented.

[Rella et al.] Clarified as follows: We have calculated the Allan standard deviation of these data (Fig. 10) in the same way as described in section 2.3. Unlike the raw data, which show increased noise after approximately 1-3 hours (Fig. 2), the Allan standard deviation of the calibrated data follows a $\tau^{(-1/2)}$ law past 100 hours of averaging, indicating that the instrument calibrated in this manner is stable over long periods of time.

L460 - 462; please consider rephrasing this - the use of English is a little colloquial in the first half of the sentence and is likely to confuse a non-English speaker.

[Rella et al.] Clarified as follows:

The impact of water vapor and carbon dioxide on the isotope ratios are non-linear in the contaminant gas concentration. In the table, we note the range of isotope ratios that captures 95% of the observed variability under varying contaminant gas conditions, as noted in the far right column of the table.

L462 - 464; this sentence reads more like an equipment manual - suggest replacing "it is recommended" with "we find that"

[Rella et al.] Corrected as follows: For the most accurate results, we conclude that the gas stream should be dried to < 0.1% mole fraction of water vapor prior to analysis, and that the variability of carbon dioxide be limited if possible.

L476; this is where section 3 should start (with 3 becoming 4 etc.)(having this breakpoint at L551 is also a possibility.)

[Rella et al.] New section inserted here.

L522; was the vehicle stopped?

[Rella et al.] The following sentence was added:

The vehicle was generally in motion during this 10 minute analysis.

L542 - 544; this sentence appears to contradict one two sentences later (L545 - 548)

[Rella et al.] The two sentences are self-consistent. The first states that the observed range of isotope ratios (for either type of well, oil or gas) is consistent with previous observations of well head gases in this basin. The second states that oil wells have a distinct signature from gas wells. No changes have been made at this time.

L542: what is C2/C1?

[Rella et al.] Ethane to methane ratio. Corrected in the text.

L561; is the effective dilution factor not a function of E_c?

[Rella et al.] The dilution factor (which is the proportionality constant that connects emission rate to concentration) does not depend on emission rate unless the emission rate is very large compared to the dilution of the atmosphere itself; this condition does not apply for small leaks that are tiny compared to the mass advective / convective motion of the atmosphere. Typically, D*E_c is < 1 x 10^-5. No changes made to the manuscript.

L571; the background here is surely the concentration of methane in the air entering, rather than the air itself

[Rella et al.] Good catch. Corrected.

L583; it should be stated here which region the study of Karion et al was located in. There's also an "of" missing.

[Rella et al.] Corrected.

L596,599; please be consistent in what you call r_s

[Rella et al.] Corrected.

L600; word missing, which will fix the grammar

[Rella et al.] Corrected.

L603; this equation is orphaned and needs text around it

[Rella et al.] Corrected.

L606; This is as subsidiary goal rather than a goal? Also put in "Uintah"

[Rella et al.] Corrected as follows:

To quantify the emission rates of oil wells relative to gas wells, we need a method for collecting a representative sample of the air in the Uintah Basin. We have designed a system that samples gas over long periods of time from the mobile lab. This gas sample is analyzed in a stationary laboratory, where careful calibration and longer measurement times can be brought to bear to improve the precision and accuracy of the measurements of δ^13 CH_4 and ethane.

L612; No vehicle in the figure!

[Rella et al.] Corrected.

L613; the word "also" should be slipped in

[Rella et al.] Corrected.

L647; on what basis then is D chosen if there are no free parameters

[Rella et al.] D is from a reference (Marrero and Mason 1972), which is now included in the text.

L653; replacing "optimizing" with "minimizing" would be clearer

[Rella et al.] Good suggestion. Corrected.

L687; these are regional values, rather than just values?

[Rella et al.] Corrected as follows:

In this way, we obtain values for CH_4, $\delta^{13}CH_4$, and ethane that correspond to these 1 km path segments.

L688; this sentence is a bit unclear - what is a functional correspondence?

[Rella et al.] You are correct, the sentence as written is unclear. It is rewritten as follows:

The reanalysis algorithm provides a functional relationship between the time axis of the recorded data to that of the reanalyzed data. Using this relationship, we associate the isotope and ethane signatures measured in the laboratory with the latitude and longitude recorded in the vehicle.

L708; spelling

[Rella et al.] Corrected.

L739-741; "The 2013 numbers" is too colloquial. But in any case the way this sentence is written raises red flags, and you might consider rephrasing.

[Rella et al.] The Niwot Ridge data is now available for this time period, and the paragraph has been rewritten to reflect this (see below). In addition, as discussed in Referee #1's comments, the Monte Carlo has been rerun with the new background values, leading to small 1-2% changes in the reported ratios. The text has been updated with the new outputs.

During the atmospheric inversion event, it is difficult to define a specific background value. For the purposes of this analysis, we assign the concentration measured at Niwot Ridge in Feb. 2013 of 1.877 ppm (Dlugokencky et al. 2015) as the background concentration. The small fraction of the concentration enhancement that is due to sources outside the Uintah Basin will then contribute to the overall uncertainty of the emissions attribution; this contribution is difficult to quantify, but is expected to be small given the large enhancements observed in this study. These sources of uncertainty in the background concentration (and isotope ratio) are captured in the Monte Carlo simulation, discussed below.

L790; A researcher can be highly confident, but a lower bound cannot!

[Rella et al.] Corrected.

Figures

light gray is not a good choice of colour for reproduction on paper - it's hardly visible in Fig 14. Consider a black dashed or dot-dashed line. The same comment applies to fig 15.

[Rella et al.] In Fig. 14, dashed and dashed-dotted lines did not represent the fast fluctuations well. Instead, the light gray color has been replaced with cyan. For figure 15, increasing the visibility of the fine gray lines (which are only present to indicate the locations of roads) clutters the figure. We have left these light gray lines in the figure as is.

Fig. 15 it needs to be made explicit that the color bar refers to delta. An inset that gives the reader an idea of how symbol area corresponds to concentration, would be very helpful

[Rella et al.] Corrected. We have indicated the maximum concentrations observed during each drive directly on the figure.

Fig 16 please explain the inset ("0201" etc.)

[Rella et al.] Corrected.