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PAPER DRAFT: Using high frequency δ^{13}C-CH$_4$ and δ^{2}H-CH$_4$ observations to constrain inverse-modelled methane emissions, whilst considering uncertain regional isotopic signatures

Alice E. Ramsden[1], Anita L. Ganesan[2], Matthew Rigby[3], Chris Rennick[4], Emmal Safi[5], Tim Arnold[5,6], Edward Chung[4,5], Alistair J. Manning[1], Dave Lowry[7], Simon O'Doherty[3], and Kieran Stanley[3]

[1]Met Office Hadley Centre, Exeter, UK
[2]School of Geographical Sciences, University of Bristol, Bristol, UK
[3]School of Chemistry, University of Bristol, Bristol, UK
[4]National Physical Laboratory, Teddington, UK
[5]School of Geosciences, University of Edinburgh, Edinburgh, UK
[6]Department of Physical Geography and Ecosystem Science, Lund University, Lund, Sweden
[7]Department of Earth Sciences, Royal Holloway, University of London, Egham, UK

Correspondence: Alice E. Ramsden (alice.ramsden@metoffice.ac.uk)

Abstract. This draft paper presents an inverse modelling method for estimating methane emissions using methane isotope delta value observations to aid with emission source attribution. The motivation for this work and the history of the use of secondary observations with methane inverse modelling is discussed, before showing how this work builds on that history. The inverse modelling method and how this has been adapted to include methane isotope delta observations and uncertain source signatures is then presented. The submitted version of this publication will include synthetic data tests to demonstrate the ability of this model to estimate UK fossil-fuel and non-fossil-fuel methane emissions, followed by a short case study using this method with new high frequency methane isotope delta value observations from the UK Heathfield observation site.

1 Introduction

In 2022, global monthly average atmospheric methane (CH$_4$) concentrations exceeded 1900 parts per billion (ppb). Since then, concentrations have continued to rise, with some of the largest annual increases on record occurring in the last 5 years (Lan et al., 2023). This continuing rise in growth rate suggests that current attempts to stabilise or reduce anthropogenic methane emissions are not effectively targeting the causes of this rise. Improved mechanisms for monitoring these attempts is required in order to reduce methane's impact on current and future climate change. Inverse modelling of emissions, as demonstrated in this work, can contribute to this process.

Methane is a key target for emissions reduction because of its short tropospheric lifetime (compared to other greenhouse gases) of approximately 10 years and relatively high 100-year global warming potential (GWP$_{100}$) of approximately 28 times that of carbon dioxide (Myhre et al., 2013). A rapid reduction in the emission of methane into the atmosphere could have a significant impact on overall climate forcing (Ganesan et al., 2019) which is required in order to meet global climate change targets, including those set out in the 2015 Paris Agreement. The Agreement aims to keep mean global temperature rise to below 2°C above pre-industrial temperatures, by requiring all signatories to plan for and regularly report on emissions targets. To aid with setting these emissions targets, the Intergovernmental Panel on Climate Change (IPCC) have produced Shared Socio-economic Pathways (SSPs) which detail various emission scenarios and their impact on future global temperature rises (Arias et al., 2021). Recent global atmospheric methane concentrations do not show any of the decline which is required to meet the more sustainable SSPs which would enable us to mitigate any further rises in global average temperature. The ongoing rise in global methane concentration shows that current emissions targets are not being met or that these targets are not stringent enough to satisfy the Paris Agreement goals. Therefore, greater focus on the monitoring of country-level methane emissions is required in order to monitor these targets and meet the lower emission scenarios (Nisbet et al., 2019).

Accurate monitoring of methane emissions, which is required in order to effectively target emissions reduction strategies, is complicated by methane's wide range of anthropogenic and natural sources. For the period 2010-2019, global average methane emissions were approximately 620 teragrams per year (Tg y^{-1}) (Saunois et al., 2025). Approximately 40% of these global emissions are from natural sources: wetlands, freshwater sources, permafrost and geological seeps. As most of the methane from these natural sources is emitted during the decomposition of organic matter by methantrophic organisms in waterlogged environments, these sources are highly seasonal and can be dependent on temperature and precipitation (Pison et al., 2013). Approximately 60% of total methane emissions are from anthropogenic sources, with agriculture and waste management (including enteric fermentation by livestock, manure management, landfills and waste water treatment) making up approximately 35% of all methane emissions. Fossil fuel production, transportation and use contributes approximately 20% of total emissions (Saunois et al., 2025). Biomass and biofuel burning also produce small levels of methane emissions.

Methods used to estimate global, regional and country-level methane emissions and their sources can be split into two main types: bottom-up studies, which model emissions processes to directly estimate sector-level surface emissions using geographic and economic data; and top-down inverse modelling studies, which improve on an initial (prior) estimate of emissions by using an atmospheric transport model to infer total surface emissions from atmospheric methane mole fraction (concentration) observations. Sector attribution in top-down estimation is challenging and often cannot be carried out using only information from methane mole fraction observations. Traditionally, a percentage of the final (posterior) total methane flux estimate is attributed to each sector, based on the percentage each source contributes to total emissions as given by a bottom-up dataset. This partitioning can be applied at various resolutions depending on the level of confidence in the spatial and temporal distributions in the underlying bottom-up dataset.

Bottom-up methods are regularly used by national governments to compile national inventories. These inventories are then reported to the United Nations Framework Convention on Climate Change as part of the Paris Agreement emissions reporting process. Top-down models have more commonly been used by the scientific community to learn about trends in global emissions (e.g. Maasakkers et al. (2019); Zhang et al. (2021)), study regional emissions and their sources (e.g. Ganesan et al. (2015); Sheng et al. (2018)) or measure emission rates from large point sources (e.g Cui et al. (2019)). More recently, top-down models have been used as independent verification of bottom-up national GHG inventories (Henne et al., 2016; Manning et al., 2021) and newer international research projects often include direct collaboration between top-down modelling scientists and national inventory teams (e.g. PARIS (2023); AVENGERS (2023)).

1.1 Using methane isotope observations for source attribution

Recent developments in top-down inverse modelling methods, including those presented in this paper, have allowed for a more detailed comparison of top-down and inventory estimates of emissions, by estimating sector-level emissions directly during the inversion. These methods often use observations of secondary trace gases (e.g. C_2H_6 or CO) or methane isotopes (e.g. δ^{13}C-CH_4 or δ^{2}H-CH_4) to aid with source attribution. Methane is created by a range of production mechanisms, some which produce distinct ratios of co-emitted gases or isotopes. The source of emission can be inferred when these characteristic ratios are detected in observations. For example, ethane is co-emitted from methane's fossil fuel sources, but there is negligible emission from methane's other biogenic sources, so observations of ethane can be used to infer a fossil fuel methane source in local methane plume studies (e.g. Mielke-Maday et al. (2019)) or regional inverse modelling (e.g. Barkley et al. (2021)). See Ramsden et al. (2022) for a more detailed summary of the use of secondary mole fraction observations in methane inverse modelling; the rest of this paper will focus on the use of methane isotope observations for source attribution in top-down modelling of methane emissions.

Methane from biogenic (agriculture, waste and wetland) sources contains a lower ratio of ^{13}C to ^{12}C than methane from thermogenic (fossil fuel) sources, so observations of this ^{13}C:^{12}C ratio can be used to infer the proportion of sources contributing to total emissions. Similarly, ^{2}H (D, deuterium):H observations can be used for source attribution because biological methane sources are more depleted in heavier hydrogen isotopes than fossil fuel methane sources. These characteristic ratios are referred to as source signatures.

Due to the greater availability of long term, low frequency δ^{13}C-CH_4 observations, these observations are most commonly used in the top-down modelling of global methane emissions to attribute trends in emissions to a source sector (e.g. Fujita et al. (2020); Milkov et al. (2020); Lan et al. (2021)). Methane isotope observations have also been used on much smaller spatial scales, to attribute methane plumes to their point source by using Keeling plot analysis to find the δ^{13}C-CH_4 isotopic signature of the local methane enhancement (France et al., 2016; Cain et al., 2017). Keeling plot analysis (and the similar Miller-Tans process) has also been used to attribute emission peaks in longer methane time series (e.g. (Menoud et al., 2021; Varga et al., 2021; Hoheisel and Schmidt, 2024)). Menoud et al. (2021) found that the use of δ^{2}H-CH_4 observations, alongside δ^{13}C-CH_4 was vital for their case study in Krakow, Poland where there was overlap between locally observed fossil fuel and microbial δ^{13}C-CH_4 source signatures that was not present in the observed δ^{2}H-CH_4. Similarly, Röckmann et al. (2016) demonstrated how new high-resolution δ^{13}C-CH_4 and δ^{2}H-CH_4 observations at Cabauw in the Netherlands could be used to constrain the source attribution of western European emissions, and found results contradicted a bottom-up model's source allocation. When using δ^{13}C-CH_4 observations, Zazzeri et al. (2017) and Saboya et al. (2021) also found top-down estimates of London's methane emissions which contradicted those from a bottom-up UK inventory. Source attribution using the isotope observations led to the conclusion that under-reported local gas leaks were responsible for the mismatch.

The above studies have all used fixed values or fixed ranges for methane isotope source signatures when comparing observed to modelled isotope values. However, these signatures can vary spatially and temporally depending on the type of methane production or composition of reaction products (Sherwood et al., 2017). More recent databases of source signatures are improving current understanding of source signatures and allowing for better constraints to be placed on these parameters in inverse models. This include databases created by Sherwood et al. (2017) and Milkov et al. (2020) for fossil fuel sources, Menoud et al. (2022) for European sources, Bakkaloglu et al. (2021) for UK landfill sources and Lowry et al. (2020) for a range of UK sources.

These developments, alongside the availability of higher frequency methane isotope observations have led to top-down methods using isotope observations that explicitly consider the uncertainty in the source signatures. Thanwerdas et al. (2022, 2024) presented a global inverse modelling method using a modified version of the Community Inversion Framework (CIF) with weekly or monthly smoothed observations of methane, δ^{13}C-CH$_4$ and δ^{2}H-CH$_4$ to optimise both global methane emissions and their associated source signatures. Drinkwater et al. (2023), using inverse methods to optimise both regional emissions and regional δ^{13}C-CH$_4$ signatures, concluded that a relative increase in tropical methane was causing the recent trend in global methane. They also found strong trends and variation in regional signatures across all regions of the globe between 2004 and 2020. Basu et al. (2022) incorporated both methane mole fraction and δ^{13}C-CH$_4$ observations into a global inversion model using the transport model TM5-4DVAR, but instead of optimising source signatures directly in the inversion as in Thanwerdas et al. (2022), they explored the impact of source signature uncertainty by running the inverse model with different sets of fixed source signatures. They found that whilst source signature uncertainty was not a significant contributor to overall posterior uncertainty in their methane flux estimates over large geographical regions, methane isotope observations had to be assimilated directly into the inverse model for it to produce posterior methane flux estimates that were consistent with atmospheric δ^{13}C-CH$_4$, suggesting that the methane-only inversion was not accurately attributing emissions to their sources without this information. Overall, previous works using isotope observations have shown strong potential for use in providing more independent top-down attribution of emissions to their source, but have also highlighted that the use of single isotope tracers may limit a model's ability to separate sources when there is overlap between source signatures or when source signatures are uncertain. This uncertainty has been considered in some cases, but often is not factored into the final flux estimates.

This paper aims to build on these previous works by presenting a novel inverse modelling method that uses new high-frequency observations of both δ^{13}C-CH$_4$ and δ^{2}H-CH$_4$ and optimises regional signatures alongside methane emissions to estimate methane emissions from fossil fuel (FF) and biogenic, non fossil fuel (non-FF) sources. This model is based on work presented in Ramsden et al. (2022) which used observations of methane and ethane mole fractions and optimised both methane emissions and ethane:methane emission ratios. The modifications made to this method to incorporate isotope delta value observations and regional signatures are presented in Section 2 alongside a more general introduction to the inverse modelling framework. The potential of this method to estimate UK fluxes using both current methane, ethane and methane isotope observations and future expansions to the UK observational network will be tested in a range of synthetic data tests. This will be followed by a short case study using the new dataset of high frequency methane isotope observations to demonstrate how this model could be used to estimate regional FF and non-FF methane emissions.

2 Methods

This section briefly introduces top-down inverse modelling of surface emissions in general terms, before discussing how this framework is adapted in this work to include secondary observations and emission ratios or source signatures.

2.1 Inverse modelling of surface emissions from multiple sectors

This top-down inverse model uses Bayesian statistical methods to infer regional surface emissions from atmospheric mole fraction (concentration) observations. An atmospheric transport model is used to link observations to surface emissions, by representing the meteorological transport of gas particles from where they were observed, back in time to where they either were emitted from the surface, or where they entered the study domain. As observed mole fractions contain both global background concentrations and enhancements from local sources, both of these quantities need to be considered when estimating regional emissions. This system can be represented by forward version of the model:

$$\mathbf{y} = \mathbf{H} \cdot \mathbf{x} + \mathbf{H}_{bc} \cdot \mathbf{x}_{bc} + \boldsymbol{\epsilon}_y \tag{1}$$

where $\mathbf{y}$ are the atmospheric observations, $\mathbf{H}$ is the combined emissions and atmospheric transport (the modelled emissions mole fraction) and $\mathbf{x}$ are the emissions scaling factors required to match these forward modelled mole fractions to the observed mole fractions. $\mathbf{H}_{bc}$ is the combined boundary conditions and atmospheric transport (the modelled boundary condition mole fraction, which represents the mole fraction entering or exiting the domain) and $\mathbf{x}_{bc}$ are the boundary condition scaling factors. ϵ_y is the combined model and measurement error.

This model can be expanded to include an estimate of emissions scaling factors for more than one sector:

$$\mathbf{y} = \begin{bmatrix} \mathbf{H}_1 & \dots & \mathbf{H}_n & \mathbf{H}_{bc} \end{bmatrix} \cdot \begin{bmatrix} \mathbf{x}_1 \\ \vdots \\ \mathbf{x}_n \\ \mathbf{x}_{bc} \end{bmatrix} + \boldsymbol{\epsilon}_y \tag{2}$$

where $\mathbf{H}_{1\dots n}$ represents the transport and emissions from each sector (or type of source).

Using a Bayesian statistical framework allows for an initial (a priori) estimate of emissions to be included in the model. In most cases, a bottom-up emissions inventory is used for this purpose. Similarly, a prior estimate of the background (boundary condition) is needed, usually estimated using a global emissions inventory.

Bayes theorem is used to infer the most likely (posterior) emissions, given the atmospheric observations and the a priori emissions and boundary conditions estimates. To ease computation, the emissions and boundary conditions priors and transport footprints are combined before the inversion, into the $\mathbf{H}$ and $\mathbf{H}_{bc}$ matrices as shown above. Posterior scaling factors of the emissions ($\mathbf{x}$) and boundary conditions ($\mathbf{x}_{bc}$), relative to their priors are then optimised by the inverse model. This allows for emissions to be solved for at a coarser resolution than the transport model and allows for easy customisation of this parameter for sensitivity and synthetic data tests.

As in previous studies (Ganesan et al. (2014); Ramsden et al. (2022)) a hierarchical form of Bayes theorem is used, which allows for secondary parameters, such as emission ratios of a gas and a tracer, regional isotope ratio signatures (from here on referred to as regional signatures) or model uncertainty to also be optimised during the inverse modelling process:

$$\rho(\mathbf{x},\mathbf{R},\boldsymbol{\theta}|\mathbf{y}) \propto \rho(\mathbf{y}|\mathbf{x},\mathbf{R},\boldsymbol{\theta}) \cdot \rho(\mathbf{x}) \cdot \rho(\mathbf{R}) \cdot \rho(\boldsymbol{\theta}). \tag{3}$$

The hierarchical model also allows for non-Gaussian probability density functions (PDFs) to be specified for all input parameters. For example, truncated Gaussian PDFs can be used for those parameters for which negative values are unrealistic or uniform PDFs can used for parameters which are not well constrained.

The most probable values of **x** (and the other variable parameters) can be found by minimising the following cost function:

$$\begin{aligned}\rho(\mathbf{x},\boldsymbol{\theta}_n|\mathbf{y}_n) \propto\ & (\mathbf{y}-\mathbf{H}\mathbf{x})^T\mathbf{Q}_y^{-1}(\mathbf{y}-\mathbf{H}\mathbf{x}) \\ & + (\mathbf{x}-\mathbf{x}_{prior})^T\mathbf{P}_x^{-1}(\mathbf{x}-\mathbf{x}_{prior}) \\ & + (\mathbf{x}_{bc}-\mathbf{x}_{bc,prior})^T\mathbf{P}_{x_{bc}}^{-1}(\mathbf{x}_{bc}-\mathbf{x}_{bc,prior}) \\ & + \sum_i^m(\boldsymbol{\theta}_i-\boldsymbol{\theta}_{prior_i})^T\mathbf{P}_{\theta_i}^{-1}(\boldsymbol{\theta}_i-\boldsymbol{\theta}_{prior_i})\end{aligned} \tag{4}$$

where $\mathbf{Q}_y$ is the model-measurement uncertainty matrix, $\mathbf{P}_x$, $\mathbf{P}_{x_{bc}}$ and $\mathbf{P}_{\theta_i}$ are the prior uncertainty matrices for the emissions, boundary conditions and any number (m) of secondary parameters ($\boldsymbol{\theta}$).

As there is no analytical solution available to minimise this cost function, an iterative MCMC process based on the Metropolis-Hastings algorithm is used. This process involves randomly sampling each variables prior PDF and calculating the probability of this sample relative to the sample of values in the previous iteration. If this new sample is more likely, it is retained or ‘accepted’ and a new sample is taken from this set of values in the next iteration. However, if the sample is less likely it is rejected and the chain returns to its previous set of values. To prevent the model from converging at local minima or maxima, some randomly selected ‘less likely’ sets of samples are also accepted. The step size used to dictate the size of the distance between old and new samples is optimised to produce a sampling acceptance ratio of approximately 30%. This optimisation is performed using a adapted version of Algorithm 4 from Andrieu and Thoms (2008).

The first 50% of samples are discarded as a ‘burn-in’ period, to reduce any impact of the model’s initial state on the posterior, and every 100^{th} value of the remaining trace is retained to form a posterior distribution. This thinning is carried out to reduce the memory size of any output files and should not have any impact on the posterior PDF if the traces are sufficiently well converged. Convergence is tested at multiple steps during the inversion, by comparing the percentage and absolute differences between the mean of the previous 40% and 20% of iterations. The percentage difference between the first and second halves of the posterior traces are also compared, to confirm convergence.

2.2 Inverse modelling with secondary isotope ratio observations

This inverse method can be adapted to include methane isotope ratio observations. Methane isotope ratio measurements are presented as a delta value: a ratio of the minor to major isotopologues relative to an international standard, typically written in permille. For observations of the two isotopologues used in this work (δ^{13}C-CH$_4$ and δD-CH$_3$D), delta values δ^{13} and δ^{2} in terms of permille units are expressed as follows:

$$\delta_i = \left(\frac{R_i}{R_{std_i}} - 1\right) \times 1000 \tag{5}$$

where i is the isotopologue and R$_{i,std}$ is the isotopic standard. For δ^{13}, R$_{13,std}$ is the Vienna PeeDee Belemnite (VPDB) standard with a value of 0.011180 and for δ^{2}, R$_{2,std}$ is the Vienna Standard Mean Ocean Water (VSMOW) with a value of 0.00015575 (Brand et al., 2019; Werner and Brand, 2001).

Equation 5 can be rearranged to give the isotopic ratio in terms of the observed delta value:

$$R_i = R_{std_i}\left(\frac{\delta_i}{1000} + 1\right) \tag{6}$$

When using delta value secondary observations, the relationship between modelled delta values and methane emissions is non-linear and is dependent on the emissions from each sector and the accompanying source signatures for each sector. Therefore, the forward model for delta values becomes:

$$\mathbf{y}_i = \mathbf{H}_s . f\left(\mathbf{x}_s, \boldsymbol{\delta}_{i,s}\right) + \mathbf{H}_{bc} . f\left(\mathbf{x}_{bc}, \boldsymbol{\delta}_{i_{bc}}\right) \tag{7}$$

where $\mathbf{y}_i$ is the modelled delta value for each delta value type (i), $\mathbf{H}_s$ are the combined transport matrices and emissions priors for each sector (s), $\mathbf{x}_s$ are the emissions scaling factors for each sector, $\delta_{i,s}$ are the source signatures for each sector, $\mathbf{H}_{bc}$ is the combined transport matrix and boundary condition prior, $\mathbf{x}_{bc}$ are the boundary condition scaling factors and $\boldsymbol{\delta}_{i_{bc}}$ are the boundary condition signatures, linking methane mole fraction boundary conditions to delta value boundary conditions.

Source signature delta values are included as the secondary variable parameter in the inversion and the relative abundance of different methane istopologues implied by these source signatures is used to produce modelled delta values, which can then be compared to the delta value observations. These modelled delta values are updated during the iterative inversion, as new values of source signatures and methane emission scaling factors are proposed. Modelled delta values are created using the following process:

1. The absolute fraction of each isotopologue (a_i) is calculated from the sample of source signature delta values using the following equations (derivations of these equations is given in detail in Appendix A):

   $$\begin{aligned} a_{^{12}\mathrm{CH}_4} &= \frac{1}{(1+R_{13}+R_2)} \\ a_{^{13}\mathrm{CH}_4} &= \frac{1}{\left(1+\frac{1}{R_{13}}+\frac{R_2}{R_{13}}\right)} \\ a_{^{12}\mathrm{CH}_3\mathrm{D}} &= \frac{1}{\left(1+\frac{1}{R_2}+\frac{R_{13}}{R_2}\right)} \end{aligned} \tag{8}$$

   where R_{13} and R_2 are the isotopic ratios derived from observations of δ^{13} and δ^2, using Equation 6.

2. Modelled mole fractions are created for each isotopologue using the forward model for total methane mole fractions and the absolute fractions for each isotopologue:

   $$\mathbf{y}_i = \begin{bmatrix} \mathbf{H}_1 & \ldots & \mathbf{H}_n & \mathbf{H}_{bc,CH_4} \end{bmatrix} \cdot \begin{bmatrix} \mathbf{a}_{i,1}\mathbf{x}_1 \\ \vdots \\ \mathbf{a}_{i,n}\mathbf{x}_n \\ \mathbf{a}_{i,bc}\mathbf{x}_{bc} \end{bmatrix} + \boldsymbol{\epsilon}_{y,i} \tag{9}$$

   where *n* is the number of emission sectors and **x** are the current states of the emission scaling factor terms.

3. The isotopic ratio is calculated from the ratio of these isotopologue mole fractions:

   $$R_{^{13}CH_4} = \frac{\mathbf{y}_{^{13}CH_4} m_{^{13}CH_4}}{\mathbf{y}_{^{12}CH_4} m_{^{12}CH_4}} \tag{10}$$

   $$R_{^{12}CH_3D} = \frac{\mathbf{y}_{^{12}CH_3D} m_{^{12}CH_3D}}{\mathbf{y}_{^{12}CH_4} m_{^{12}CH_4}} \tag{11}$$

   The combined transport matrix and emissions priors used in step 4 (**H**) contain a conversion from mass to molar terms using the molar mass of methane. As the absolute mole fractions of each isotopologue are being considered here, the conversion from total methane molar mass to isotopologue molar mass needs to be included (m_i/m_{CH_4}).

4. These ratios are converted into delta values using the isotopic delta value equation (Equation 5).

Other than these extra steps used to create modelled delta values, the MCMC process including variable source signatures is similar to that with emission ratio scaling factors. The delta value source signatures have their own prior PDFs, proposed values are sampled and their probability is tested against both the emissions and source signature prior PDFs, and modelled delta values from these source signatures are tested against the delta value observations.

3 Further work

Testing of this method is underway and includes synthetic data tests and a case study with Boreas methane isotope delta value observations at Heathfield, in the UK. This work is currently in progress and the final submitted version of this paper will include the setup, results and conclusions from the model testing and UK case study.

Appendix A: Calculating isotopologue absolute fractions

This method assumes that there are only three methane isotopologues present in the atmosphere: $^{12}\mathrm{CH}_4$, $^{13}\mathrm{CH}_4$ and $^{12}\mathrm{CH}_3\mathrm{D}$. The total methane mole fraction can be found by summing the contributions from each isotopologue:

$$\mathrm{CH}_4 = {}^{12}\mathrm{CH}_4 + {}^{13}\mathrm{CH}_4 + {}^{12}\mathrm{CH}_3\mathrm{D} \tag{A1}$$

The absolute mole fraction of each isotopologue can be found by considering the ratio of isotopologues given by delta value observations and substituting this information into Equation A1.

Methane isotope observations are presented as delta values:

$$\delta^i = \left(\frac{R_i}{R_{std_i}} - 1\right) \times 1000 \tag{A2}$$

where R_i for the two sets of methane isotope observations used in this work are:

$$R_{13} = \frac{^{13}CH_4}{^{12}CH_4} \tag{A3}$$

$$R_2 = \frac{^{12}CH_3D}{^{12}CH_4} \tag{A4}$$

To find the absolute fraction of $^{12}\mathrm{CH}_4$:

Equations A3 and A4 are rearranged then substituted into A1:

$$\begin{aligned}
{}^{13}\mathrm{CH}_4 &= {}^{12}\mathrm{CH}_4 R_{13} \\
{}^{12}\mathrm{CH}_3\mathrm{D} &= {}^{12}\mathrm{CH}_4 R_2 \\
\mathrm{CH}_4 &= {}^{12}\mathrm{CH}_4 + {}^{12}\mathrm{CH}_4 R_{13} + {}^{12}\mathrm{CH}_4 R_2 \\
\mathrm{CH}_4 &= {}^{12}\mathrm{CH}_4\,(1 + R_{13} + R_2) \\
{}^{12}\mathrm{CH}_4 &= \frac{\mathrm{CH}_4}{(1 + R_{13} + R_2)} \qquad \text{(A5)}
\end{aligned}$$

Similarly for $^{13}\mathrm{CH_4}$ and $^{12}\mathrm{CH_3D}$:

$$
\begin{aligned}
{}^{12}\mathrm{CH_4} &= \frac{{}^{13}\mathrm{CH_4}}{R_{13}} \\
{}^{12}\mathrm{CH_3D} &= {}^{12}\mathrm{CH_4}R_2 \\
\mathrm{CH_4} &= \frac{{}^{13}\mathrm{CH_4}}{R_{13}} + {}^{13}\mathrm{CH_4} + {}^{12}\mathrm{CH_4}R_2 \\
\mathrm{CH_4} &= \frac{{}^{13}\mathrm{CH_4}}{R_{13}} + {}^{13}\mathrm{CH_4} + \frac{{}^{13}\mathrm{CH_4}}{R_{13}}R_2 \\
\mathrm{CH_4} &= {}^{13}\mathrm{CH_4}\left(\frac{1}{R_{13}} + 1 + \frac{R_2}{R_{13}}\right) \\
{}^{13}\mathrm{CH_4} &= \frac{\mathrm{CH_4}}{\left(1 + \frac{1}{R_{13}} + \frac{R_2}{R_{13}}\right)} \qquad \text{(A6)}
\end{aligned}
$$

$$
\begin{aligned}
{}^{13}\mathrm{CH_4} &= {}^{12}\mathrm{CH_4}R_{13} \\
{}^{12}\mathrm{CH_4} &= \frac{{}^{12}\mathrm{CH_3D}}{R_2} \\
\mathrm{CH_4} &= \frac{{}^{12}\mathrm{CH_3D}}{R_2} + {}^{12}\mathrm{CH_4}R_{13} + {}^{12}\mathrm{CH_3D} \\
\mathrm{CH_4} &= \frac{{}^{12}\mathrm{CH_3D}}{R_2} + \frac{{}^{12}\mathrm{CH_3D}}{R_2}R_{13} + {}^{12}\mathrm{CH_3D} \\
\mathrm{CH_4} &= \frac{{}^{12}\mathrm{CH_3D}}{\frac{1}{R_2} + \frac{R_{13}}{R_2} + 1} \\
{}^{12}\mathrm{CH_3D} &= \frac{\mathrm{CH_4}}{\left(1 + \frac{1}{R_2} + \frac{R_{13}}{R_2}\right)} \qquad \text{(A7)}
\end{aligned}
$$

These terms can then used to produce modelled atmospheric delta values using surface source signature estimates and the isotropic ratio equation A2.

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