

Dear Fortunat,

Thank you for your assistance in the publication process of our manuscript.

This is a point-by-point reply to all comments made by referees and in short comments, as well as your own. Below these, we have listed additional corrections and changes to the manuscript that became necessary during the revisions but are not directly related to external comments. A revised manuscript and a copy that shows a mark-up of all changes have also been submitted. Please note that we have added Gordon Brailsford as additional author, as discussed before.

Sincerely,

Hinrich

We thank all referees and commenters for the time taken to evaluate our study and for the helpful comments and suggestions. Below we address each criticism individually. Some points were brought up by several referees and commenters; in those cases our responses and changes to the manuscript may not be repeated for every single one, but all concerns have been addressed. Referees' comments are shown in italics. Our response is in regular font. Quotes from the manuscript are in quotation marks; bold type indicates new wording in the manuscript.

15

Editor's comment:

<Personally, I missed a discussion on the potential variability in the signature of $d13C(CH_4)$ of different sources as well as of the sinks and how such variations may affect your results. For example, I could imagine that variations in the C3 versus C4 origin of organic source material could have an influence on wetland $d13C(CH_4)$ emissions.>

Unfortunately, we had missed this comment in the acceptance letter for discussion and therefore did not address it with the earlier revisions. Variability in sinks is now been discussed in response to comments from other referees and commenters. We have now added the following passage in the introduction on the variability of source isotopic signatures and C3/C4 wetlands:

“Varying contributions from wetlands dominated by C₃ and C₄ plants, which differ in the $\delta^{13}C_{CH_4}$ of their emissions, may be part of the ENSO-CH₄ signal or work to obscure it if controlled by other drivers. In general, we assume that $\delta^{13}C_{CH_4}$ of the various emission sources has not changed over the ~35 yr period of our study. Although such changes, correlated to atmospheric CO₂ mole fractions, have been reported to occur over centuries to millennia in ice core studies (Möller et al., 2013), they are likely negligible over the short duration and >20% CO₂-change of our study period.”

30

Referee #1:

<The manuscript (...) provides additional evidence that emissions sources that have high interannual-variability, i.e., wetlands and fire, are unlikely to be the dominant cause of sustained emissions.>

This is an important point that we now state in the conclusions:

Conclusions: “**The longer the atmospheric [CH₄] and δ¹³CH₄ trends persist, the less probable are processes that impact IAV and short-lived cyclical events like ENSO as the driver.**”

5 <First, I recommend the authors revisit the title and modify to be more specific than just ‘methane cycle’ because this implies the authors were looking at methane emissions, but rather the authors investigated atmospheric concentrations. I would prefer a title along the lines of “Limited impact of El Niño – Southern Oscillation on the atmospheric methane growth anomalies”.>

10 We have changed the title to: “**Limited impact of El Niño – Southern Oscillation on variability and atmospheric growth rate of methane**”. This addresses the concerns voiced by the reviewer and also reflects that the revised manuscript also discusses sink dynamics.

15 <Second, the Introduction could be clearer to reflect that the authors are motivated by understanding atmospheric methane concentration anomalies rather than anomalies in emissions. The previous studies linking methane emissions to ENSO as a key driver are not in question, but currently the Introduction mixes a little the emissions and concentrations anomalies making the reader have to work to clarify this.>

20 We have clarified the scope and focus of the study. Please note that changes in response to SC2 are also relevant to this point. Resulting changes to the manuscript are as follows:

Abstract: “Here, we test the impact of ENSO on **atmospheric** CH₄ in a correlation analysis.”

Introduction: “Attributing recent changes in the methane budget, **and the associated impact on its growth rate**, to specific natural or anthropogenic causes is essential for climate change mitigation.”

25 Introduction: “We conduct correlation analyses between ENSO variability and [CH₄], as well as δ¹³CH₄ records to quantify **how much ENSO anomalies in emissions and sinks affect atmospheric CH₄**. Specifically, we explore how much of the year-to-year variability in methane **levels** can be attributed to ENSO...”

Introduction: “The aim is to detect the impact of ENSO on atmospheric CH₄ **levels** on various spatial scales.”

30 Conclusions: “Further identification of these **processes** is necessary to inform climate change mitigation policies and climate projections.”

35 Further, following suggestions from both reviewers and an additional short comment, the revised manuscript also discusses sink dynamics. The relationship between the latter and variability in emissions is now clarified in the introduction. For more details on the treatment of sink dynamics please see the response to SC1.

40 <In Table 1, I assume the lag time is in months, so 54 is a 54 month lag? If so, many are longer than 12 months, which is contrary to the statement in Section 5.2 that says most are shorter than a year.>

We have clarified that lag times in the tables are reported in months, e.g. by including this information in the table captions. The statement that detrended time series at SMO generally have their highest correlations at lag times of less than one year

holds true (the only exceptions are for EMI, as well as a few cases for SMO det-gro). We have modified the text to make it clearer that the short lag times only apply to SMO detrended data series.

Section 5.1.2.: "...our analysis therefore allows for lag times of up to 5 years in monthly increments in the calculations and reports the maximum r^2 and lag time (**in months**) for a given ENSO-[CH₄]/ $\delta^{13}\text{CH}_4$ /HCN combination."

5 Section 5.2.: "Methane mixing ratios show correlations with ENSO of r^2 -values up to 0.36 at SMO, but only for detrended time series (Table 1). [...] **For SMO detrended [CH₄] series**, lag times are fairly consistent across the various ENSO indices and generally shorter than 1 year. For other [CH₄] records at SMO and ASC the **highest** correlations are $r^2 < 0.23$ **and have** lags of over 3 years.

10 *<I am skeptical of such long lags, it is difficult to judge whether shorter lags were close in terms of significance to the longer lag times because these numbers are not presented.>*

We agree that allowing for lags up to 5 years leads to questionable results. However, it is difficult to define a cut-off for lags. The current presentation provides an upper limit for ENSO influence. Given that the latter is found to be low, this represents
15 a conservative estimate. For most dependent time series, there are cases of comparable r^2 -values for lags both longer and shorter than 3 years. Using a 3-year cut-off for lags therefore does not really affect the conclusions. Nevertheless, for specific r^2 -values with lags over 3 years that are mentioned in results and discussion, we now also report the corresponding highest r^2 for lags of less than 3 years. We have revised sections 5.1.2. and 5.2. to address this point.

Section 5.1.2.: "A lag time between ENSO forcing and detection of resulting $\delta^{13}\text{CH}_4$ or HCN variability at the measurement
20 site, (or in the global average) is likely, due to a **variety of factors that may lead to lags of unknown length and some of which may be cumulative: e.g., hydrology, plant growth and decay, microbial response, seasonal triggers for methanogenesis or burning, as well as atmospheric chemistry, mixing, and transport between source regions and sampling sites. Therefore, it is difficult to define a cut-off for lags.** Literature estimates of **specific lags range...**"

Section 5.2.: "Although the analysis provides r^2 -values for lags up to 60 months (Tables 2-4), we consider it likely that
25 lags of >3 years indicate spurious correlations given that individual ENSO events last 1-2 years and global atmospheric mixing times are on order of 1 year. Therefore, we also report the highest r^2 for lags <3 years in the following sections. For other cases with lags >3 years in Tables 2-4, the highest relevant r^2 -value is lower than the reported value, where the latter places an upper limit on the influence of ENSO."

Section 5.2.: "For other [CH₄] records at SMO and ASC the highest correlations are $r^2 < 0.23$ and have lags of over 3 years
30 ($r^2 < 0.19$ for lags <3 years). The global running mean [CH₄] time series shows $r^2 = 0.24$ (lag: 4.5 years; $r^2 = 0.04$ for lag <3 years) with the SOI running mean for the period 1998-2016.

Referee #2, Joe Melton:

We thank Joe Melton for the constructive and helpful comments and suggestions. Below we address each criticism individually. Please note that some points were brought up by several referees and commenters; please see our other responses for additional information and changes to the manuscript. Referees' comments are bracketed as follows: <>. Our response is
5 in regular font. Quotes from the manuscript are in quotation marks.

Response to main comments:

<1. I didn't see a discussion of significance level for the correlation coefficients. Without one I have trouble understanding if a value of 0.25 is significant or not. Is there a reason that was not done? Otherwise, while all of the r^2 are low, it might help understand what are just noise and what is representing a true signal.>

10

We have marked all correlation results for the Spearman ranks that are not statistically robust at a level of $p > 0.01$ with grey backgrounds in the results tables. This applies only to results with very low r -squared values and therefore does not affect the interpretation. The exception are correlations of global $\delta^{13}\text{CH}_4$ with ENSO, which all have $p > 0.05$. These correlations did not reveal a strong ENSO impact either, so removing them from the interpretation does not alter the findings. The highest
15 correlations have been revised downwards by a slight margin.

The resulting changes in the manuscript are as follows:

Abstract: "We find that at most **36%** of the variability in $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ is attributable to ENSO, but only for detrended records in the Southern tropics. Trend-bearing records from the Southern tropics, as well as all studied hemispheric and global records show a minor impact of ENSO, i.e. <**24%** of variability explained."

20

Section 5.2.: "**P-values for the Spearman ranks indicate that all results for $r^2 > 0.1$ are significant ($p < 0.001$), with the exception of global $\delta^{13}\text{CH}_4$ correlations, where no p-values below 0.05 occur.**"

Section 5.2.: "**None of the global $\delta^{13}\text{CH}_4$ series produced statistically robust correlations with ENSO; all p-values were higher than 0.05. The following findings are therefore not relevant for further interpretation.** The highest correlation is between global detrended $\delta^{13}\text{CH}_4$ and SOI monthly means with $r^2 = 0.37$."

25

Section 5.2.: "**The lack of statistical robustness for global $\delta^{13}\text{CH}_4$ -ENSO correlations may stem from the different resolution of the two sets of time series. In this case, the southern hemispheric record from BHD may represent the extra-tropical impact of ENSO on $\delta^{13}\text{CH}_4$.**"

30

<2. There is relatively little discussion on possible changes in the main sink of methane - hydroxyl radicals. The dynamics of this sink has been highlighted in recent studies (McNorton et al. 2016, Turner et al. 2017, Rigby et al. 2017). Is it possible that ENSO would have an impact upon local concentrations of OH? Is it a safe assumption to assume a constant sink strength? Given the power of this sink, and its recent importance (at least in the studies just mentioned) perhaps it is worthwhile to give more consideration to how a changing sink due to ENSO could impact upon the methane cycle. Or at least expand the discussion of the sink to demonstrate why a constant assumption is valid.>

35

A detailed discussion of OH-dynamics has been included in the revised manuscript, for details please see replies to SC1 by Alex Turner.

Response to minor comments:

<- *What about using the GFED4(s) burned area product to investigate changes in burning? It would have the advantage that it is global and extends further back in time than the HCN record. However the caveat is that burned area is not the same as C emitted as CH₄, however it might be a reasonable test since they would be closely related. A recent study highlighted the continual decrease in burnt area over the course of the record (Andela et al. 2017) which then should have some impact upon the methane cycle and perhaps can be used to tease out any ENSO influence.*>

10 Evidence for an influence of ENSO on earlier versions of the GFED data set has been presented by Van der Werf et al. (2006). Although GFED now covers a longer period, we see limited value in repeating the analysis. The biomass burning proxy HCN as an atmospheric tracer provides a more direct comparison with the methane records (e.g., both are subject to atmospheric transport and mixing). Given that the focus of our study is the overall impact of ENSO on methane dynamics, with biomass burning emissions as a piece of the puzzle, we prefer to maintain a clear scope of the work.

15 <- *Can we have a table with the various tests laid out (det-nom, det-gro, nominal mm, nominal run, etc.)? It is difficult to keep them all in the head and then interpret some very busy figures.*>

We have included a new Table 1 that provides a list and description of all data sets.

<- *p. 2 line 30 - What is aggregate source here?*>

20 We have changed the wording to read:

Introduction: “Biogenic methanogenesis in wetlands discriminates strongly against ¹³C and creates methane that is ¹³C-depleted ($\delta^{13}\text{C} = -58\text{‰}$ for tropical wetlands) relative to the plant precursor material ($\delta^{13}\text{C}$ of -12‰ to -28‰) and to the **combined total of global emissions** ($\delta^{13}\text{C} \sim -53.5\text{‰}$).”

<- *Author contributions - there is no E.D author.*>

25 We have corrected the author contributions. Before the original submission we mutually agreed that Ed Dlugokencky’s contribution would be more appropriately reflected in an acknowledgement rather than through co-authorship.

<- *Data avail - raw data is little use. Best approach would be to make the actual analysis available.*>

We are now stating that all data products and time series used in the study are available from the corresponding author.

30

Interactive comment SCI:

We thank Alex Turner for the time taken to evaluate our study and for the helpful comments and suggestions. Below we address each criticism individually. Please note that some points were brought up by several referees and commenters; please see our other responses for additional information and changes to the manuscript. Referees’ comments are bracketed as follows:

<>. Our response is in regular font. Quotes from the manuscript are in quotation marks.

<The manuscript argues for a limited role of ENSO on the methane cycle; however, the manuscript makes little mention of two important factors that impact atmospheric methane and are strongly influenced by ENSO: (1) atmospheric transport and (2) loss via hydroxyl. These factors seem particularly pertinent to a discussion of the role of ENSO on the methane cycle. There have been a number of recent papers on these two topics in the last two years that the authors seem to have overlooked. Specifically, McNorton et al. (2016), Turner et al. (2017), and Rigby et al. (2017) showed how changes in the methane loss via oxidation by hydroxyl was an important factor in the interpretation of methane trends.>

These papers are relevant for a complete picture of the methane cycle in the introduction and conclusion. We have added references in these sections. In contrast, for the core question of our study they are not relevant, just as anthropogenic sources like fossil-fuel methane are not. We note that a recent OH-reconstruction (Naus et al., 2018) finds that sink trends may be less relevant than modelled by Turner et al. (2017) and Rigby et al. (2017).

Changes to the manuscript are as follows:

Introduction: **“Considering recent reconstructions of methane’s dominant atmospheric sink, i.e. the hydroxyl radical OH, we consider it likely that increasing emissions contribute to (Rigby et al., 2017), if not dominate (Naus et al., 2018), the [CH₄] rise. If so, the methane source type that varied can be investigated...”**

Introduction: **“Changes in OH have also been suggested as partial or dominant drivers in recent CH₄ trends, both for the onset of the 1999-2006 plateau (McNorton et al., 2016; Schaefer et al., 2016) and for the post-2007 [CH₄] increase (Rigby et al., 2017; Turner et al., 2017).”**

Conclusions: **“Changes in removal rates via OH have been suggested as an additional (Rigby et al., 2017) or alternative (Turner et al., 2017) driver of the increase, but recent work suggests that sink impacts are not dominant (Naus et al., 2018).”**

<More directly related to ENSO, Corbett et al. (2017) showed the influence of ENSO on the spatial distribution of methane via changes in atmospheric transport... >

The role of transport is relevant for the tropical time series. We have included a discussion of the findings of Corbett et al. (2017) as a possible explanation why ENSO signals are smaller than may have been anticipated. However, we also note that the observed anomalies in mid-tropospheric [CH₄] are inconsistent with the patterns expected from emission changes. On hemispheric or global scales transport processes are unlikely to play a strong role, given the short mixing time of methane relative to its atmospheric turn-over.

Changes to the manuscript are as follows:

Section 5.3.7.: **“Corbett et al. (2017) show that during La Niña events high surface temperatures over the Western Pacific lead to upward transport over the Indonesian region (a CH₄ source area from wetlands and rice paddies) and negative CH₄ anomalies in the mid-troposphere (tropical surface air with relatively low [CH₄] replaces air from the Northern Hemisphere with higher [CH₄]). This mechanism would dampen the signal of higher La Niña emissions in**

surface records like SMO and ASC. However, the corresponding El Niño anomalies in mid-tropospheric CH₄ over the Central Pacific are smaller. This indicates that Central Pacific surface air, where there are no CH₄ sources, is closer in [CH₄] to mid-tropospheric levels than surface air from the Western Pacific. Unless there were strong longitudinal differences in mid-tropospheric [CH₄], this is inconsistent with a scenario where high concentrations of CH₄ are generated over the Western Pacific in La Niñas but transported upwards and away from the surface stations used in this study. On hemispheric or global scales transport processes are unlikely to play a strong role, given the short mixing time of methane relative to its atmospheric turn-over.”

<...while Turner et al. (2018) showed how ENSO can strongly influence the methane lifetime.>

The findings of Turner et al. (2018) on tropical OH-dynamics during ENSO events are very relevant to this study. They are now laid out briefly in the introduction and are discussed in depth in the revised section 5.3.7. In short, OH-dynamics are expected to provide a negative feedback on methane concentration signals from ENSO emissions but a positive feedback on the stable isotope signal. This offers an additional explanation why ENSO impacts on methane growth rates is less than has been suggested in some studies. It would also make δ¹³CH₄ a more sensitive tracer for ENSO impacts but our records do not show the expected δ¹³CH₄ signals.

Changes to the manuscript that result from this discussion are as follows:

Abstract: “Dynamics of the removal by hydroxyl may counteract the variation in emissions, but the expected isotope signal is not evident.”

Introduction: “A chemistry climate model suggests that ENSO modulates tropical OH (where hydroxyl levels are highest) via changes in NO_x production through lightning, ozone availability and specific humidity, as well as emissions of reactive carbon (Turner et al., 2018). Resulting changes in methane removal could create their own signal in atmospheric records of [CH₄] and δ¹³CH₄. They could also either reinforce or dampen the emission impacts discussed above.”

Section 5.3.7.: “The low correlations of [CH₄] and δ¹³CH₄ with ENSO rule out a dominant role for ENSO triggered sink changes in atmospheric methane records. Removal processes could lead to either amplification or dampening of source signals. Higher emissions of methane and CO from biomass burning will draw down OH and weaken the sink. Emission factors from fires for CO are between 10 and 30-fold higher than for CH₄ (Van der Werf et al., 2017), so that the biomass burning dynamics dominate the source of reactive carbon, leaving less OH during El Niños and more during La Niñas to draw down CH₄. This would provide a negative feedback for the emissions [CH₄]-signal from ENSO forcing. In contrast, the feedback on the ENSO emissions δ¹³CH₄-signal would be positive due to varying enrichment of ¹³C-methane through sink fractionation (less removal leads to less ¹³C-enrichment of relatively ¹³C-depleted wetland emissions during La Niñas; more removal increases the ¹³C-enrichment from biomass burning emissions during El Niños further). In addition to the reactive carbon effect, (Turner et al., 2018) found a further OH increase during La Niñas due to higher lightning rates with NO_x production. Turner et al. (2018) could attribute 17% of OH variability

that is forced by climate cycles (rather than emissions of other atmospheric compounds) to ENSO. This is a minor part of the variability, but in consequence, the dampening effect on [CH₄] and the reinforcing feedback on δ¹³CH₄ would be even larger. In our correlation results these sink impacts are not apparent, as the [CH₄] correlations for the tropical stations are higher than δ¹³CH₄ correlations (Tables 1 and 2). Nevertheless, the OH-dynamics provide a possible explanation for the limited ENSO impact on [CH₄] variability and trends. They also make δ¹³CH₄ a conservative proxy for the influence that ENSO exerts on tropical methane. Whether ENSO has less influence on CH₄ emissions than assumed or whether such an impact is overwhelmed by **atmospheric removal** or other CH₄ cycle processes,...

Conclusions: “As δ¹³CH₄ is subject to a mutually reinforcing signal from ENSO suppression of wetland emissions and enhancement of biomass burning CH₄ (or vice versa), **as well as positive feedbacks from OH-dynamics**, it is particularly suited to study the role of ENSO in the CH₄ cycle.

Conclusions: “**Counteracting OH-dynamics are expected to further dampen any influence ENSO may have on methane growth rates.**”

Conclusions: “Our results do not rule out that ENSO influences CH₄ emissions from wetlands and biomass burning through temperature, enhanced precipitation or droughts in key regions, but any such impacts are overwhelmed **by OH-dynamics** or other source and sink processes.”

Interactive comment SC2:

We thank Zhen Zhang for the time taken to evaluate our study and for the helpful comments and suggestions. Below we address each criticism individually. Please note that some points were brought up by several referees and commenters; please see our other responses for additional information and changes to the manuscript. Referees’ comments are bracketed as follows: <>. Our response is in regular font. Quotes from the manuscript are in quotation marks.

<I found the title is a bit misleading as it sounds like ENSO has limited impacts on CH₄ sources and sinks, which is not supported by previous studies.>

We have clarified the goal and scope of the study in the introduction. We have also changed the title following suggestions from this comment and other referees. Relevant changes to the title and manuscript are as follows:

Title: “Limited impact of El Niño – Southern Oscillation on **variability and growth rate of atmospheric methane**”

Abstract: “Here, we test the impact of ENSO on **atmospheric** CH₄ in a correlation analysis.”

Abstract: “It is possible that other processes obscure the ENSO signal, which itself indicates a minor influence of the latter on **global** CH₄ emissions.”

Introduction: “We conduct correlation analyses between ENSO variability and [CH₄], as well as δ¹³CH₄ records to quantify **how much** ENSO anomalies in emissions and sinks affect atmospheric CH₄. Specifically, we explore how much of the

year-to-year variability in methane **levels** can be attributed to ENSO and how large the ENSO-CH₄ signal is in dependence of latitude.”

5 Conclusions: “Further identification of these **processes** is necessary to inform climate change mitigation policies and climate projections.”

<-The response of CH₄ concentration to natural CH₄ sources could be weak during weak/moderate ENSO events and the methane sources from anthropogenic activities could be dominating.>

10 The goal of our study is to find out whether the cumulative effect of ENSO events on the methane cycle is strong enough to drive observed trends in atmospheric methane (see response above). This does indeed mean that other processes may dominate. In fact, this is the main finding of the paper and has been discussed explicitly in the original submission, e.g., in Section 5.3.7., which is titled “Role of other methane cycle processes” and in the conclusions. Relevant passages include: “...or other
15 processes in the CH₄-cycle obscure the ENSO impacts.”; “ENSO could affect CH₄ emissions from tropical wetlands and biomass burning as predicted by Hodson et al. (2011) and van der Werf et al. (2006), respectively, but the resulting isotopic signal is overwhelmed by other components of the CH₄ cycle. Such influences could be other sources (particularly anthropogenic ones), variability in atmospheric transport or changes in CH₄ sink processes.”; and “Our results do not rule out that ENSO influences CH₄ emissions from wetlands and biomass burning through temperature, enhanced precipitation or
20 droughts in key regions, but any such impacts are overwhelmed **by OH-dynamics or** other source and sink processes.”

<Also, the general assumption of lower CH₄ during El Ninos seems to be controversial to the observations in some El Nino event (e.g. 1997/98).>

25 The focus of our study is if the impact of ENSO on atmospheric CH₄ is persistent throughout our records. Working with general assumption of lower CH₄ during El Ninos, consistent with the findings in the latest paper authored by the commenter, is therefore a valid approach. The variance in expression between different ENSO events is discussed in the original submission, section 5.3.4. concluding with the sentence: “Depending on the strength and geographical expression of the climate anomaly, ENSO may thus cause regional or global emission anomalies that are opposite to the expected pattern.”

30 *<I feel it would be very helpful if the authors could add an additional analysis to maximize the signals by focusing on strong ENSO events.>*

35 Attribution of signals to specific ENSO events is better approached with different methods and data sets than used in our study. Several recent publications on this topic are cited in the original submission, e.g., Pandey et al. (2017). It is also not the question we are trying to answer. Strong events may cause larger signals, but they are also rarer and more sporadic, so that their impact on our time series is limited. This is discussed in section 5.3.3. Another point on this suggestion is that the commenter’s last publication identifies only four strong El Ninos, two of which occur before the start of our tropical d13CH₄ and HCN records.
40 It is impossible to find general patterns with such a small sample size. We therefore prefer to maintain the scope of our study. Please note that the original submission already provided the following finding on strong El Ninos and their impact on the correlations:

“The full BHD record for 1992-2016 gives very similar results as the 1998-2016 subset used for comparison with the other
45 stations (as discussed above). However, the shorter subset for 1998-2014 produces larger Pearson r²-values (0.26 for running means and SOI), and for 2001-2014 we find Pearson r²-values up to 0.38 (growth rate correlated to EMI). These shorter data sets omit the strong El Niño events of 1998 and/or 2015-16, which could have been expected to have a strong influence on methane emissions and consequently δ¹³CH₄.”

5 <-For C13-CH4, the authors assume that a detectable change in C13-CH4 during ENSO
should be observed if ENSO has significant impacts on wetland and biomass burning
given that the suppression of wetland and enhanced biomass burning act in the same
direction on C13-CH4. I wonder if this signal can be detected without removing noises
from other factors like atmospheric transport, local OH, and other biogenic sources
which are also influenced by climate conditions (e.g. landfills and agricultural sources,
10 which have similar C13-signature as wetlands and also respond to changing rainfall
and temperature).>

This comment goes back to the one above on interfering signals from other processes (see response above). While the
individual arguments are all correct, they do not invalidate our point from the original submission: “If ENSO is invoked as a
main cause of recent trends in [CH₄] and δ¹³CH₄ this should be manifested in sizeable correlations.” We have added a note in
15 the introduction regarding the commenter’s point that some anthropogenic sources could reinforce the wetland anomalies:
Introduction: **“Several anthropogenic sources are subject to the same ENSO forcing and are expected to vary in concert
with wetlands (e.g., rice agriculture, possibly livestock).”**

20 <In addition, the growth of wetland CH4 emissions during El Ninos is more complex than previous thoughts, Zhang et al.,
(2018) suggest that wetland CH4 emissions were suppressed at the early stage of El Nino but the wetland CH4 growth rate is
in the rising phase at the later stage of El Ninos.>

It is unclear how much mixing and transport would smooth out the evolving signal from an El Nino event in our atmospheric
records. Nevertheless, the sequence has the potential to cause patterns in monthly growth rates (strong positive anomalies) that
25 are in contradiction with the general pattern of reduced overall emissions. We have noted this in sections 5.2. and 5.3.4.:

**“The highest values are from (detrended) growth rates, which can be more indicative of dynamics within an ENSO
event, rather than its overall emissions impact (Zhang et al., 2018).”**

30 **“Zhang et al., (2018) report an evolving response of wetland emissions to El Niños, where an initial reduction due to
decreased wetland extend is counteracted by increased microbial activity under higher temperatures during the later
stages of the event.”**

35 <Given that the peak of CH4 growth for wetland and biomass burning occur differently, this could weaken the net impact on
the C13-CH4 signal.>

This was explicitly stated in the original submission (Section 5.3.1.):
40 “It is possible that biomass burning and wetland CH₄ production have different response times to ENSO forcing, which would
weaken their cumulative impact on δ¹³CH₄.”

<- Zhang et al., (2018) suggests that wetland CH4 emission could have a step increase
of ~9 Tg CH4/yr for the period of 2007-2014 compared to 2000-2006. Could this affect
some of the authors’ conclusions?>

45 The finding indeed affects certain conclusions, although with the caveat that Melton et al. (2014) show that different wetland
models show a large range in modelled emissions and do not necessarily agree in the simulated trends. Agreement of the model
ensemble would be needed for a robust conclusion. We have added the following passages:

Section 5.3.6.: “**Other wetland variability may have contributed to the rise (Zhang et al., 2018); given the range in wetland model output (Melton et al., 2014) this stands to be confirmed by ensemble runs.**”

Section 6.: “Our results **do** not rule out that wetland production is a contributor to the post-2007 [CH₄]-rise if driven by environmental controls other than ENSO. **This is suggested by a modelled increase in wetland CH₄ production between the periods 2000-2006 and 2006-2014, although with the limited confidence of a single wetland emissions model (Zhang et al., 2018).**”

Please note that we have added additional passages - or amended existing ones - informed by the study of Zhang et al. (2018) with regards to various points:

10 Section 5.3.2.: “One explanation for the lower combined wetland-pyrogeic $\delta^{13}\text{CH}_4$ signal is low sensitivity of wetland CH₄ production to ENSO events. **This is consistent with r^2 -values of 0.12-0.26 between modelled wetland methane emissions (using different climate data sets as drivers) and MEI as reported by Zhang et al., (2018).**”

Section 5.3.3.: “Previous findings that modelled tropical (Zhu et al., 2015) **and global (Zhang et al., 2018)** wetland CH₄ emissions can explain at most 25% **and 14%, respectively**, of the variation in atmospheric methane growth rates therefore agree with our results that ENSO exerts only a minor control on **atmospheric CH₄**.” (note that the previous version read “... ENSO exerts only a minor control on global CH₄ emissions”)

Section 5.3.4.: **(Zhang et al., 2018) report an evolving response of wetland emissions to El Niños, where an initial reduction due to decreased wetland extend is counteracted by increased microbial activity under higher temperatures during the later stages of the event.** A complex response of wetland CH₄ production is not only seen in models, however.

20 Conclusions: “There is evidence for additional methane emissions from agriculture **(Wolf et al., 2017) ...**”

Additional changes to the manuscript:

Gordon Brailsford (NIWA) has been included in the author list to reflect his contributions to the BHD measurements. The author contributions have been updated accordingly.

25 We have added an acknowledgement to D. Lowe, who started the BHD time series.

We have corrected throughout the manuscript an error that [CH₄] is reported as mixing ratio, while it is mole fraction.

We have included additional references on technical aspects of the [CH₄] measurements at BHD:

“In addition, we use data measured at the NZ National Institute of Water and Atmospheric Research (NIWA) from BHD in NZ (41.41°S, 174.87°E; 1992-2017) **(Lowe et al., 1991). Both data sets are on the same international scale (Dlugokencky et al., 2005), although for the presented analysis internal consistency of the time series is the relevant criterion; inter-laboratory offsets do not affect the findings.**”

We have corrected a sentence in the introduction where the terms “enriched” and depleted” had been mixed up:

The ENSO impact on $\delta^{13}\text{CH}_4$ should be more pronounced than the one on [CH₄], because changes in wetland and biomass burning emissions combine to **enrich** atmospheric CH₄ in ¹³C during El Niños and **deplete** it during La Niñas.

Other clarifications and additions:

Introduction: “**Isotope** studies **suggest** that biogenic methane sources make either a dominant (Schaefer et al., 2016; Nisbet et al., 2016) or a strong (Worden et al., 2017) contribution to the recent [CH₄] rise.”

Conclusions: “**We review literature estimates of ENSO-driven emissions and find them too small and sporadic to**
5 **account for the post-2007 rise.**”

In addition, several typos have been corrected.

Limited impact of El Niño – Southern Oscillation on ~~the~~variability and growth rate of atmospheric methane~~-cycle~~

Hinrich Schaefer¹, Dan Smale¹, Sylvia E. Nichol¹, Tony M. Bromley¹, Gordon W. Brailsford¹, Ross J. Martin¹, Rowena Moss¹, Sylvia Englund Michel², James W. C. White²

¹Climate and Atmosphere Centre, National Institute of Water and Atmospheric Research, Wellington, 6021, New Zealand

²Institute of Arctic and Alpine Research, Boulder, CO, USA.

Correspondence to: Hinrich Schaefer (Hinrich.Schaefer@niwa.co.nz)

10 **2. Abstract.** The El Niño – Southern Oscillation (ENSO) has been suggested as a strong forcing in the methane cycle and as
a driver of recent trends in global atmospheric methane ~~levels~~ mole fractions [CH₄]. Such a sensitivity of the global CH₄ budget
to climate events would have important repercussions for climate change mitigation strategies and the accuracy of projections
for future greenhouse forcing. Here, we test the impact of ENSO on ~~the~~atmospheric CH₄ ~~cycle~~ in a correlation analysis. We
use local and global records of ~~methane mixing ratio~~ [CH₄], as well as stable carbon isotopic records of atmospheric CH₄
15 ($\delta^{13}\text{CH}_4$), which are particularly sensitive to the combined ENSO effects on CH₄ production from wetlands and biomass
burning. We use a variety of nominal, smoothed and detrended time series including growth rate records. We find that at most
38.36% of the variability in [CH₄] and $\delta^{13}\text{CH}_4$ is attributable to ENSO, but only for detrended records in the Southern tropics.
Trend-bearing records from the Southern tropics, as well as all studied hemispheric and global records show a minor impact
of ENSO, i.e. <25.24% of variability explained. Additional analyses using hydrogen cyanide (HCN) records show a detectable
20 ENSO influence on biomass burning (up to 51%-55%), suggesting that it is wetland CH₄ production that responds less to
ENSO than previously suggested. Dynamics of the removal by hydroxyl likely counteract the variation in emissions, but the
expected isotope signal is not evident. It is possible that other processes obscure the ENSO signal, which itself indicates a
minor influence of the latter on global CH₄ emissions. Trends like the recent rise in atmospheric [CH₄] can therefore not be
attributed to ENSO. This leaves anthropogenic methane sources as the likely driver, which must be mitigated to reduce
25 anthropogenic climate change.

3. Keywords: Time series analyses, isotopes, climate variability, climate change, methane, stable isotope analysis [more]

4. Introduction

Attributing recent changes in the methane budget, and the associated impact on its growth rate, to specific natural or anthropogenic causes is essential for climate change mitigation. The impact of climatic variability on methane emissions is particularly important to assess the potential for CH₄ release under future climate scenarios (e.g., from permafrost and wetland environments, as well as gas hydrates) in a reinforcing feedback. Atmospheric methane mixing ratios/mole fractions [CH₄] have increased by 140% over preindustrial levels (MacFarling Meure et al., 2006). The associated increase in radiative forcing makes CH₄ the second-most important anthropogenic greenhouse gas (Shindell et al., 2009). The long-term [CH₄] increase until the late 1990s can be attributed to increasing emissions from fossil fuel production (Ferretti et al., 2005; Schaefer et al., 2016), as well as sources from agriculture (enteric fermentation in livestock, rice production), waste management and anthropogenic burning (van Aardenne et al., 2001; Saunio et al., 2016). After a plateau in the early 2000s, [CH₄] has been rising again since 2007. The signature of stable carbon isotopes in atmospheric methane ($\delta^{13}\text{C}_{\text{CH}_4}$) is Considering recent reconstructions of methane's dominant atmospheric sink, i.e. the hydroxyl radical OH, we consider it likely that increasing emissions contribute to (Rigby et al., 2017), if not dominate (Naus et al., 2018), the [CH₄] rise. If so, the methane source type that varied can be investigated with measurements of stable carbon isotope ratios in atmospheric methane ($\delta^{13}\text{C}_{\text{CH}_4}$). The latter are influenced by the relative source contributions from ¹³C-depleted biogenic, ¹³C-rich pyrogenic, and thermogenic methane with intermediate $\delta^{13}\text{C}$. Studies show Isotope studies suggest that biogenic methane sources make either a dominant (Schaefer et al., 2016; Nisbet et al., 2016) or a strong (Worden et al., 2017) contribution to the recent [CH₄] rise. Biogenic methane comes predominantly from wetlands and agriculture. Schaefer et al, (2016) suggested agriculture as the more likely cause, primarily because satellite data place the increased emissions in Southeast Asia, India and China (Houweling et al., 2014). However, this geographic footprint from an inversion of satellite data is also consistent with fluxes from one particular wetland emissions model (Houweling et al., 2014). Other studies also assume a stronger role of wetlands due to drier conditions during the plateau years (Bousquet et al., 2006) and higher wetland emissions afterwards, which are attributed to a switch to predominant La Niña conditions around 2007 (Bousquet et al., 2011; Nisbet et al., 2016). La Niña is the cold phase of El Niño – Southern Oscillation (ENSO) cycles, which have a strong impact on precipitation anomalies in tropical regions (Ropelewski and Halpert, 1987; Lyon and Barnston, 2005) (Fig.1) that are key source areas for methane production from wetlands and biomass burning (Kirschke et al., 2013). ENSO impacts are strongest in the tropics, generally from December to February. During El Niño (La Niña) events in the December to February period, it tends to be drier (wetter) in the Indonesian region, north-east Brazil and south-eastern Africa, whereas it tends to be wetter (drier) in the southern USA and Mexico, eastern China and Taiwan, and east-central Africa (Fig. 1). During El Niño (La Niña) events in the June to August period, it tends to be drier (wetter) in the Indonesian region, central America and India.

The generally drier conditions during an El Niño phase Niños suppress global wetland emissions in models by up to 19 Tg/yr in the 1990s (Hodson et al., 2011). Several anthropogenic sources are subject to the same ENSO forcing and are expected to vary in concert with wetlands (e.g., rice agriculture, possibly livestock). At the same time, dry El Niño phases enhance CH₄

emissions from both natural and anthropogenic biomass burning (van der Werf et al., 2006). Wet La Niña conditions have the opposite effect; summed across the globe they increase wetland emissions and lower biomass burning CH₄. As tropical wetland fluxes are considerably larger than biomass burning emissions (Saunois et al., 2017), the expected net effect is a lower [CH₄] growth rate caused by El Niño conditions and a higher one due to La Niñas. The ENSO impact on δ¹³CH₄ should be more pronounced than the one on [CH₄], because changes in wetland and biomass burning emissions combine to ~~deplete~~enrich atmospheric CH₄ in ¹³C during El Niños and ~~enrich~~deplete it during La Niñas. Biogenic methanogenesis in wetlands discriminates strongly against ¹³C and creates methane that is ¹³C-depleted (δ¹³C = -58‰ for tropical wetlands) relative to the plant precursor material (δ¹³C of -12‰ to -28‰) and to the ~~aggregate source~~combined total of global emissions (δ¹³C ~ -53.5‰). In contrast, during burning the isotope ratios of the precursor plant material are essentially conserved and lead to δ¹³C ~ -22‰ for CH₄ emissions from fires (Schwietzke et al., 2016). The simultaneous suppression of ¹³C-depleted wetland CH₄ and enhancement of very ¹³C-rich pyrogenic emissions (and vice versa) act in the same direction on the δ¹³CH₄ of the combined source. The latter should be detectable in atmospheric δ¹³CH₄ records if the impact of ENSO on the CH₄ cycle is sufficiently large, as is predicted by the emission anomalies in wetland emission models (Hodson et al., 2011), reconstructed from satellite observations of burned area (van der Werf et al., 2010), and observed through variability in hydrogen cyanide (HCN) (Pumphrey et al., 2018), which is an indicator of biomass burning.

Varying contributions from wetlands dominated by C₃ and C₄ plants, which differ in the δ¹³CH₄ of their emissions, may be part of the ENSO-CH₄ signal or work to obscure it if controlled by other drivers. In general, we assume that δ¹³CH₄ of the various emission sources has not changed over the ~35 yr period of our study. Although such changes, correlated to atmospheric CO₂ mole fractions, have been reported to occur over centuries to millennia in ice core studies (Möller et al., 2013), they are likely negligible over the short duration and >20% CO₂-change of our study period.

Changes in OH have also been suggested as partial or dominant drivers in recent CH₄ trends, both for the onset of the 1999-2006 plateau (McNorton et al., 2016; Schaefer et al., 2016) and for the post-2007 [CH₄] increase (Rigby et al., 2017; Turner et al., 2017). A chemistry climate model suggests that ENSO modulates tropical OH (where hydroxyl levels are highest) via changes in NO_x production through lightning, ozone availability and specific humidity, as well as emissions of reactive carbon (Turner et al., 2018). Resulting changes in methane removal could create their own signal in atmospheric records of [CH₄] and δ¹³CH₄. They could also either reinforce or dampen the emission impacts discussed above.

We conduct correlation analyses between ENSO variability and [CH₄], as well as δ¹³CH₄ records to quantify ~~the strength of the impact of how much~~ ENSO ~~on the~~ anomalies in emissions and sinks affect atmospheric CH₄ ~~cycle~~. Specifically, we explore how much of the year-to-year variability in ~~the~~ atmospheric methane ~~budget~~ can be attributed to ENSO and how large the ENSO-CH₄ signal is in dependence of latitude. We test if recent trends in methane growth rate can be attributed to wetland emissions controlled by ENSO dynamics or if agricultural sources are more likely drivers. ENSO is quantified by four different indices, which are based on ocean temperature, sea level pressure gradients and a multivariate combination. [CH₄] and δ¹³CH₄ time series from four different locations were used, two from stations in the Southern tropics (Samoa, SMO, and Ascension Island, ASC), the Southern mid-latitudes (Baring Head, NZ; BHD) taken as representative of the Southern hemisphere, and

global average time series of [CH₄] and δ¹³CH₄ calculated from a network of global stations (Dlugokencky et al., 2011; Schaefer et al., 2016). We also investigate ENSO's impact on HCN data measured in Lauder, NZ (LAU), to quantify the biomass burning contribution separately. The aim is to detect the impact of ENSO on ~~the atmospheric CH₄ budget~~ on various spatial scales.

5 5.1. Methods

5.1.1. Data

For access to all data sets used in this study see Sect. 10

5.1.1.1. ENSO indices

We used four different indices in our analysis to cover various climatic effects of the ENSO cycle (Figs. 2A and 3A). The Southern Oscillation Index (SOI) is calculated from the gradient in mean sea-level pressure observations at Tahiti and Darwin, Australia (Troup, 1965). Further information on the SOI is given by (Horel and Wallace, 1981; Trenberth, 1976). The Ocean Niño Index (ONI) uses sea surface temperature (SST) anomalies in the eastern Pacific Niño 3.4. region (5°N-5°S, 120-170°W), which show smaller intra-seasonal variability than pressure and are further smoothed by using 3-month running means (Barnston et al., 1997; Kousky and Higgins, 2007).

The El Niño Modoki Index (EMI) is based on SST anomalies in the central Pacific (Ashok et al., 2007) rather than the eastern Pacific (the canonical El Niño). Events with the largest SST anomalies in the Modoki region show differences in the climate teleconnections to canonical El Niño events. The tropical precipitation differences are modest, but large differences in tropospheric circulation and wind anomalies (Yeh et al., 2009) can produce large extra-tropical differences in precipitation and temperature. The EMI has also been shown to be a significant predictor of tropical atmospheric ozone variations (Xie et al., 2014).

Variability in both atmospheric pressure and SST anomalies informs the Multivariate ENSO Index (MEI) (Wolter and Timlin, 1993; Wolter and Timlin, 1998). The various indices correlate highly with each other ($r^2 = 0.85$ and higher), except the EMI (r^2 between 0.33 and 0.52 for correlations with SOI, ONI and MEI), which deviates from the others during the strong 1997-198 El Niño event. Excluding the latter brings the correlation to r^2 between 0.74 and 0.79.

An ENSO index based on precipitation data, the ESPI, (Curtis and Adler, 2000) correlates very highly with the MEI, the ONI, and the SOI (r^2 of 0.902, 0.909, and 0.839, respectively). Therefore, we did not conduct separate calculations for the ESPI.

5.1.1.2. [CH₄] time series

The [CH₄] time series used in this study are from the Global Monitoring Division of the National Oceanic and Atmospheric Administration - Earth System Research Laboratory (NOAA ESRL) Carbon Cycle Cooperative Global Air Sampling Network (Dlugokencky et al., 2017). These data include records from SMO (latitude 14.24°S, longitude 170.57°W) and ASC (7.92°S, 14.42°W) as well as global averages calculated by smoothing background data temporally and zonally; all with coverage from 1983-2017. In addition, we use data measured at the NZ National Institute of Water and Atmospheric Research (NIWA) from BHD in NZ (41.41°S, 174.87°E; 1992-2017) (Lowe et al., 1991). Both data sets are on the same international scale (Dlugokencky et al., 2005), although for the presented analysis internal consistency of the time series is the relevant criterion; inter-laboratory offsets do not affect the findings. The individual time series (Figs. 2B-E) show seasonal cycles, inter-annual variability (IAV) and long-term trends. To investigate ENSO effects on these different time scales we derived the following seven records from the individual measurements at each station: (Table 1). First, the nominal monthly mean values to capture the full variability in the data (“nom”). Second, 12-month running means to represent IAV and trends (“run”). Third, monthly resolved growth rate defined as the difference between the following 12 months and the preceding 12 months (“gro”). Fourth, a residual (“res”) as calculated by seasonal trend analysis by Loess (STL, Cleveland et al., 1990). The seasonal window was set at 120 months, which forces a uniform seasonal cycle over the duration of the record. The residual therefore represents IAV in the expression of the seasonal cycle as well as other short-term anomalies. Fifth, sixth, and seventh: detrended time series where the STL trend component is subtracted from the monthly means with subsequent determination of detrended monthly means, 12-month running means, and growth rate (“det-nom”, “det-run”, “det-gro”).

20

5.1.1.3. δ¹³CH₄ time series

The δ¹³CH₄ time series used in this study were measured at three different laboratories, i.e., the Institute of Arctic and Alpine Research (INSTAAR), USA; the Institute for Environmental Physics (IUP) at Heidelberg University, Germany; and at NIWA. Details of the analytical methods are given by Schaefer et al. (2016) and references therein. All values are based on measured ¹³C/¹²C ratios and are reported in the standard δ-notation $\delta^{13}\text{C} = (\text{R}_{\text{sample}}/\text{R}_{\text{standard}} - 1) * 1000\%$ as per mille (‰) values where the reference standard is Vienna PeeDee Belemnite. Records at SMO (1998-2016) and ASC (2000-2016) are measured at INSTAAR. The BHD record (1992-2016) is based on measurements at INSTAAR and NIWA. An annually averaged global δ¹³CH₄ time series was established by Schaefer et al. (2016) based on data from INSTAAR, NIWA and IUP. In this analysis, we use the measurements covering 1992-2016 (Fig. 3C). For the global δ¹³CH₄ data set we conducted the analysis for the nominal annual means (“nom”) and growth rate, i.e. difference between two subsequent yearly values (“gro”). We also detrended the time series by subtracting linear trends for the sub-periods 1992-1999 and 2007-2016 (“det”) and then calculated

30

a detrended growth rate (“det-gro”). For the single-station $\delta^{13}\text{CH}_4$ records of BHD, ASC, and SMO we derived the same seven records as described for the $[\text{CH}_4]$ data (Fig. 3D-F).

5.1.1.4. HCN time series

HCN retrievals were computed from mid-infrared solar spectra measured at LAU (45.04°S, 169.68°E) as part of the Network for the Detection for Atmospheric Composition Change (NDACC). The time series has been described by Zeng et al. (2012), but the data used here are from updated retrievals using the improved SFIT4 algorithm (NDACC, 2014). The HCN data show strong seasonality that is even more pronounced in the updated retrievals. Zeng et al. (2012) found a significant negative trend for 1997-2009 and attributed it to variations in biomass burning. A similar deseasonalised trend is apparent in the updated record. HCN values are here reported as Petamolecules cm^{-2} . Measurements cover the period 1998-2017 when combined for two different instruments with a change-over point in 2000. We conducted our analyses for total column values (0-100 km). The latter signal is dominated by the tropospheric burden as measured in the 0-12 km height partial column; the correlation between total and tropospheric HCN is $r^2 = 0.997$. In addition, we investigated whether the stratospheric HCN burden is differently impacted by ENSO. To that end, we used the 12-100 km partial column, which holds ~22% of the total HCN burden. This layer shows lower correlation with the total column record ($r^2 = 0.45$).

Analogous to the monthly resolved methane records we constructed monthly means, 12-month running means, growth rates and STL residuals for the total column and stratospheric HCN data (Fig. 3B). No detrended records other than STL residual were considered.

5.1.2. Analysis

We conducted correlation analyses between the time series of a chosen ENSO index and either a $[\text{CH}_4]$, $\delta^{13}\text{CH}_4$, or HCN record as the dependent variable. The degree of correlation is quantified by the square (r^2 -value) of the Pearson correlation coefficient or, alternatively, of the Spearman ranking coefficient. The Pearson coefficient is more commonly used, but it assumes linear relationships between the variables and may underestimate nonlinear correlations. We therefore also used the Spearman rank, which does not require linearity. Note that not all correlation combinations were tested using both coefficients.

A lag time between ENSO forcing and detection of resulting $\delta^{13}\text{CH}_4$ or HCN variability at the measurement site, (or in the global average) is likely, due to ~~delayed reactions~~ a variety of factors that may lead to lags of unknown length and some of which may be cumulative: e.g., hydrology, plant growth and decay, microbial response, seasonal triggers for methanogenesis or burning ~~to environmental changes~~, as well as atmospheric chemistry, mixing, and transport ~~and mixing~~ between source regions and sampling sites. Therefore, it is difficult to define a cut-off for lags. Literature estimates of ~~such specific~~ lags range from days (Chamberlain et al., 2016) to 7 months (Zhang et al., 2018; Zhu et al., 2017), not counting atmospheric transport. Given ENSO variability with a periodicity of 2-7 years (McPhaden et al., 2006), our analysis therefore allows for lag times of up to 5 years in monthly increments in the calculations and reports the maximum r^2 and lag time (in months) for a given ENSO-

[CH₄]/δ¹³CH₄/HCN combination. We conducted the analysis for all permutations of the four ENSO indices as monthly means and their 12-month running means as well as the [CH₄], δ¹³CH₄, and HCN data products listed in Sect. 2.1 and Table 1. For all [CH₄], δ¹³CH₄, and HCN parameters we used the period 1998-2016, except for ASC where data are available only from late 2000. Using the same period for all time series avoids differing correlation results due to varying data coverage. The period includes the strong El Niños of 1998 and 2015, as well as the strong La Niñas of 1999, 2007 and 2010. We also calculated correlations for the period 1983-2016 ([CH₄] of SMO, ASC and global) and 1992-2016 (δ¹³CH₄ at BHD and global).

5.2. Results

Most combinations have r^2 -values <0.1 when comparing one dependent data set to the different ENSO time series (Tables ~~4-32-4~~). In the following, we only summarise results for the highest r^2 for each dependent time series (across all the nominal, smoothed and detrended records for a station). Given that Pearson coefficient and Spearman rank give comparable results (Tables ~~2 and 3~~), ~~we quote the Spearman results unless otherwise mentioned~~ 3 and 4, ~~we quote the Spearman results unless otherwise mentioned~~. P-values for the Spearman ranks indicate that all results for $r^2 > 0.1$ are significant ($p < 0.001$), except for global δ¹³CH₄ correlations, where no p-values below 0.05 occur. Although the analysis provides r^2 -values for lags up to 60 months (Tables 2-4), we consider it likely that lags of >3 years indicate spurious correlations given that individual ENSO events last 1-2 years and global atmospheric mixing times are on order of 1 year. Therefore, we also report the highest r^2 for lags <3 years in the following sections. For other cases with lags >3 years in Tables 2-4, the highest relevant r^2 -value is lower than the reported value, where the latter places an upper limit on the influence of ENSO.

Methane ~~mixing ratios~~ mole fractions show correlations with ENSO of r^2 -values up to 0.36 at SMO, but only for detrended time series (Table 1). ~~Here~~ The highest values are from (detrended) growth rates, which can be more indicative of dynamics within an ENSO event, rather than its overall emissions impact (Zhang et al., 2018). For SMO detrended [CH₄] series, lag times are fairly consistent across the various ENSO indices and generally shorter than 1 year. For other [CH₄] records at SMO and ASC the highest correlations are $r^2 < 0.23$ ~~with~~ and have lags of over 3 years- ($r^2 < 0.19$ for lags < 3 years). The global running mean [CH₄] time series shows $r^2 = 0.24$ (lag: 4.5 years; $r^2 = 0.04$ for lag < 3 years) with the SOI running mean for the period 1998-2016. However, for the full length of available data, as well as all BHD records, all correlations are below $r^2 = 0.20$, with lag times that are variable, extremely short (zero or 1 month) or over 3 years.

The highest correlations are between HCN running means for total column, as well as stratospheric growth rates, and 12-month running mean ENSO records (Table 2). Here, ENSO accounts for 30%-51% of the observed variability, depending on the ENSO index. For both total and stratospheric HCN, lag times for maximum correlation are generally shorter than one year and are consistent (≤ 6 months difference) between the various ENSO indices, with exception of the EMI.

The $\delta^{13}\text{CH}_4$ records from the stations SMO, ASC, and BHD all have r^2 -values below 0.2324 (Table 23). Variability in lag times between different ENSO indices for the same dependent record is generally high.

~~For~~None of the global $\delta^{13}\text{CH}_4$ series produced statistically robust correlations with ENSO; all p-values were higher than 0.05.

5 The following findings are therefore not relevant for further interpretation. The highest correlation is between global detrended $\delta^{13}\text{CH}_4$, 37% of variability can be attributed to and SOI monthly means with $r^2=0.37$. Global $\delta^{13}\text{CH}_4$, is the only parameter where ENSO monthly means produce higher correlations than the smoothed (12-month running mean) record. Because the correlation calculation between annual $\delta^{13}\text{CH}_4$ and ENSO monthly means is specific for the month-of-year, this finding indicates that global $\delta^{13}\text{CH}_4$ is more sensitive to the seasonality of ENSO than its IAV. The actual ENSO influence on global

10 $\delta^{13}\text{CH}_4$ is shown through correlation with running ENSO indices, which is highest between nominal $\delta^{13}\text{CH}_4$ values and SOI with Pearson $r^2 = 0.25$ for 1998-2016. For the period 1992-2016 this value drops to Pearson $r^2 = 0.20$. The lack of statistical robustness for global $\delta^{13}\text{CH}_4$ -ENSO correlations may stem from the different resolution of the two sets of time series. In this case, the southern hemispheric record from BHD may represent the extra-tropical impact of ENSO on $\delta^{13}\text{CH}_4$.

15 The full BHD record for 1992-2016 gives very similar results as the 1998-2016 subset used for comparison with the other stations (as discussed above). However, the shorter subset for 1998-2014 produces larger Pearson r^2 -values (0.26 for running means and SOI), and for 2001-2014 we find Pearson r^2 -values up to 0.38 (growth rate correlated to EMI). These shorter data sets omit the strong El Niño events of 1998 and/or 2015-16, which could have been expected to have a strong influence on methane emissions and consequently $\delta^{13}\text{CH}_4$.

20

For none of the stations (including global average) did the detrended $\delta^{13}\text{CH}_4$ time series (incl. STL residuals) produce a markedly stronger correlation with ENSO than any of the other data series from that station. This is remarkable because ENSO can be expected to have more influence on IAV than on the long-term trends, which are quite pronounced.

25 5.3. Discussion

5.3.1. General causes and caveats for correlations of $[\text{CH}_4]$, $\delta^{13}\text{CH}_4$, and HCN with ENSO

Detected correlations between ENSO indices and $[\text{CH}_4]/\delta^{13}\text{CH}_4/\text{HCN}$ by themselves do not prove a causal relationship. However, the underlying mechanisms for a potential forcing have been presented by van der Werf et al. (2006) for biomass burning and by Hodson et al. (2011) for wetland CH_4 production. Accordingly, a correlation analysis is useful to quantify an

30 upper limit of variability in the CH_4 cycle attributable to ENSO. Because ENSO simultaneously suppresses wetland CH_4 that is more ^{13}C -depleted than the cumulative methane source and enhances pyrogenic CH_4 that is more ^{13}C -enriched (or vice versa), the two influences partly cancel for the combined emission rates, i.e. their impact on $[\text{CH}_4]$. However, they reinforce

each other's impact on total source $\delta^{13}\text{CH}_4$. It is possible that biomass burning and wetland CH_4 production have different response times to ENSO forcing, which would weaken their cumulative impact on $\delta^{13}\text{CH}_4$. Similarly, longer atmospheric residence time of CH_4 (~9 years, Prather et al., 2012) over HCN (~3 months, Li et al., 2000) and a smaller relative portion of ENSO-sensitive emissions in the global methane source may lead to dampening effects in the $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ variability and hence lower correlation with ENSO indices compared to HCN. The available records for HCN and $\delta^{13}\text{CH}_4$ from ASC and SMO cover only a small number of ENSO events, which could affect the results. However, when analysing subperiods of global and BHD $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ records, we find larger correlations for shorter periods, particularly when strong ENSO events are excluded. This shows that the results are not biased against the detection of ENSO influences because records are too short. We also note that all stations measure background air, they are set up to detect broad spatial and temporal trends and not specific emission events such as an ENSO triggered plume. However, if ENSO is invoked as a main cause of recent trends in $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ (Nisbet et al., 2016) this should be manifested in sizeable correlations.

5.3.2. Contrasting correlation patterns for $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ versus HCN

In all $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ records, ENSO cycles explain about one third of the variability in detrended records and less than one quarter in others. This is true even for the Southern tropics, where ENSO has strong climatic impacts and where the majority of low-latitude wetland emissions and of biomass burning emissions originate (Kirschke et al., 2013). Correlations found for ASC and SMO, which represent this latitude band in our study, exceed those for the Southern mid-latitudes or the global record only by a limited margin and only for detrended records. Further, inconsistent lag times, lags of more than three years, and higher correlation coefficients for the exclusion of major ENSO events point to spurious correlations.

In contrast, we find a prominent influence of ENSO on the biomass burning proxy HCN. ENSO impacts on HCN have been reported before, e.g., by Pumphrey et al. (2018), who observe suppression of HCN levels during La Niña events and enhancement during El Niños, particularly in equatorial Asia. That study found a rather confined geographical impact of El Niño events with strongly enhanced HCN emissions around Malaysia, Indonesia, and Papua New Guinea, as well as generally rapid transport eastward and to the stratosphere. We speculate that the fast, upward transport (although not observed for all El Niño events) explains why stratosphere growth rates are the most sensitive data set to ENSO. For the total column, the HCN burden is concentrated in lower tropospheric levels and may be subjected to more mixing of different air parcels. According to the results of Pumphrey et al. (2018), data from LAU in the Southern mid-latitudes are outside the region of the strongest HCN signal. This is also evident in the zonal mean HCN climatologies of (Sheese et al., 2017). Yet, ENSO accounts for up to 51% of the variability in our biomass burning proxy record. ~~That~~ One explanation for the lower combined wetland-pyrogenic $\delta^{13}\text{CH}_4$ signal is lower sensitivity of wetland CH_4 production to ENSO events. This is low, or that consistent with r^2 -values of 0.12-0.26 between modelled wetland methane emissions (using different climate data sets as drivers) and MEI as reported by Zhang et al. (2018). Alternatively, other processes in the CH_4 -cycle obscure the ENSO impacts.

5.3.3. Impact of ENSO on methane emission rates

In a correlation analysis by Zhu et al. (2017), ENSO explained 49% of IAV in modelled tropical wetland CH₄ emissions. This is far higher than the combined effect with biomass burning on $\delta^{13}\text{CH}_4$ in this study and therefore seems to be an overestimate. Even so, the magnitude of the modelled emission changes is 6 Tg/yr at most. The modelling study of Hodson et al. (2011) finds slightly larger anomalies in global wetland emissions due to ENSO with mean reductions of -9 ± 3 Tg/yr and mean gains of $+8\pm 4$ Tg/yr for El Niño and La Niña events, respectively. Pandey et al. (2017) found in a comprehensive inversion study that the net effect of the strong 2011 La Niña on tropical and northern extratropical CH₄ emissions was a global increase of +6.6 Tg/yr. The wetland emission anomalies are expected to be partly compensated by changes in biomass burning that are of opposite sign. We are not aware of studies that quantify biomass burning anomalies for specific ENSO events. Assuming that ENSO is the main control of biomass burning emissions of CH₄, the IAV in the GFED data (van der Werf et al., 2010) may serve as an indication for possible ENSO impacts. In that case, the standard deviation of 2.4 Tg/yr for 1998-2014 would approximate the average impact, with maximum anomalies of up to 4 Tg/yr. We use these numbers together with results from Hodson et al. (2011) in the following proof-of-concept discussions. The combined wetland – biomass burning anomalies are ~6 Tg/yr for average ENSO events and ~8 Tg/yr for extreme ones; restricted to 1-2 yearlong individual events. This is well short of the sustained increase after 2007 when yearly emissions were ~20 Tg higher than during the 1999-2006 plateau period and the 9 Tg/yr reduction during the 1990s. ~~A previous finding of Zhu (Schaefer et al. (2015), 2016). Previous findings~~ that ~~simulated modelled~~ tropical (Zhu et al., 2015) and global (Zhang et al., 2018) wetland CH₄ emissions can explain at most 25% and 14%, respectively, of the variation in atmospheric methane growth rates ~~(possibly overestimated for the same reasons as the results of Zhu et al. (2017), see above)~~ therefore agrees agree with our results that ENSO exerts only a minor control on global atmospheric CH₄ emissions.

5.3.4. Process based understanding of ENSO impact on wetlands

A major contribution of ENSO to the recent [CH₄] increase is inconsistent with independent assessments of wetland response, as shown above, but our findings do not detect any clear minor contribution of ENSO to [CH₄] and $\delta^{13}\text{CH}_4$ timeseries, either. Several reasons may explain the lack of correlation, where we assume that wetlands respond less than proposed. The main ENSO forcing on tropical wetland CH₄ production is thought to be via wetland extent, which is driven by precipitation (Hodson et al., 2011; Holmes et al., 2015; in contrast Zhu et al., 2017, find temperature to be dominant). However, a case study in the Eastern Amazon finds that precipitation changes explain only 21% of wetland CH₄ emission variance during the wet season and 7% over the whole year (Basso et al., 2016). The lack of a direct link between precipitation and wetland CH₄ production is also evident in the large range in output from various wetland models even when forced with the same meteorological conditions (Melton et al., 2013), although the disagreement between models could also be due to an incomplete understanding of influences on the wetland cycle other than precipitation (Turetsky et al., 2014; Bridgman et al., 2013; Parker et al., 2018). Zhang et al. (2018) report an evolving response of wetland emissions to El Niños, where an initial reduction due to decreased

wetland extend is counteracted by increased microbial activity under higher temperatures during the later stages of the event.

A complex response of wetland CH₄ production is not only seen in models, however. The inversion study of Pandey et al. (2017) found a global increase of +6.6 Tg/yr for the strong 2011 La Niña, but a reduction by -6.1 Tg/yr during the 2012 weak La Niña. Similarly, Liu et al. (2017) found that El Niño conditions produced opposing weather forcing and carbon cycle responses between various tropical regions, as well as differing ones between the ~~2011~~1998 and 2015 events. Another example of this is flooding in the Amazon region during La Niña events, while flooding in the wetlands of the Paraná region occurs during El Niños (Parker et al., 2018). Depending on the strength and geographical expression of the climate anomaly, ENSO may thus cause regional or global emission anomalies that are opposite to the expected pattern.

10 5.3.5. Evaluating the consistency of ENSO impacts throughout the record

The atmospheric [CH₄] history shows global emission reductions in the 1990s and increases after 2007 (Schaefer et al., 2016). This would be consistent with ENSO forcing of the methane cycle where the 1990s were dominated by drier El- Niño periods, whereas the recent years of predominant La Niña conditions were wetter. Given that the magnitude of the low-latitude wetland CH₄ source exceeds pyrogenic emissions rates, the expected emissions history would qualitatively match atmospheric trends.

15 Also, for a short period between 2008 and 2011 Schaefer et al. (2016) observed the activation of CH₄ emissions with an extremely ¹³C-depleted cumulative δ¹³CH₄ (~-75‰). Such a value on the global scale is hard to match by a single source type. The cumulative effect of wetland enhancement and fire suppression forced by the 2008 La Niña event would provide an excellent explanation. However, the isotopic signal of the emissions reductions in the 1990s should be similar if ENSO forcing was the cause. In contrast, Schaefer et al. (2016) found that the “lost emissions” during that period are quite ¹³C-rich and rather
20 indicate a reduction in fossil fuel methane. An alternative interpretation of these isotope trends by Rice et al. (2016) requires simultaneous reductions of pyrogenic and biogenic emissions, which is also inconsistent with the expected ENSO forcing. A consistent match between ENSO phases and global δ¹³CH₄ is therefore neither evident in the dominant δ¹³CH₄ trends nor in the correlation analysis presented in this study.

25 5.3.6. Using isotopes to attribute emission changes

The impact of an ENSO emissions “perturbation” (i.e. the combined emissions anomaly of an event) on atmospheric δ¹³CH₄ can be assessed in isotope mass balance calculations according to:

$$S_{\text{total}} * \delta_{\text{total}} = S_1 * \delta_1 + S_2 * \delta_2 + S_3 * \delta_3 \quad (1)$$

Where, for a given source, S and δ represent emission rate and δ¹³CH₄, respectively (note that for scenarios discussed here S
30 may be negative, i.e., a reduction in emissions). Using generic isotope source signatures for biogenic, fossil fuel and pyrogenic methane emissions from Schwietzke et al. (2016), we find that the average La Niña perturbations proposed in section 4.3. have an effective δ¹³CH₄ of -79‰, with -83‰ for extreme ones. As expected, the combined isotope leverage of wetland enhancement and fire reductions on the global source is strong, equalling the leverage of much larger source anomalies (20

Tg/yr) with lower $\delta^{13}\text{CH}_4$ of $\sim -60\text{‰}$ after 2007 as calculated by Schaefer et al. (2016). In addition to the assumed 6 Tg/yr ENSO perturbation, another ~ 14 Tg/yr of emissions with $\delta^{13}\text{CH}_4 = -52\text{‰}$ would be necessary to produce the observed $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ trends. The isotope mass balance then shows that the non-ENSO additional emissions are a roughly equal mix of fossil fuel and biogenic methane. Noting that the assumption that all years after 2007 experienced average La Niña conditions is unrealistic, these findings therefore show ~~conservatively~~ the following three points: (i) ENSO effects alone cannot explain the post-2007 $[\text{CH}_4]$ -rise. (ii) There was an increase in ~~other~~-biogenic sources in addition to ENSO driven wetland ~~emissions anomalies~~. Other wetland variability may have contributed to the rise (Zhang et al., 2018); given the range in wetland model output (Melton et al., 2014) this stands to be confirmed by ensemble runs. In the absence of boreal emission increases (Sweeney et al., 2016), the only biogenic source large enough to accommodate ~~such the required~~ changes is agriculture (Saunio et al., 2016). (iii) Any ENSO-driven reduction in biomass burning after 2007 allows for, or requires, growing fossil fuel emissions. The latter has recently been proposed by Worden et al. (2017), who reconstructed larger biomass burning reductions after 2007 than recorded by GFED, although without assigning the reductions to ENSO or other causes.

5.3.7. Role of other methane cycle processes

There is an alternative explanation for the lack of correlation between ENSO and the methane records. ENSO could affect CH_4 emissions from tropical wetlands and biomass burning as predicted by Hodson et al. (2011) and van der Werf et al. (2006), respectively, but the resulting isotopic signal is overwhelmed by other components of the CH_4 cycle. Such influences could be other sources (particularly anthropogenic ones), variability in atmospheric transport or changes in CH_4 sink processes. A stronger ENSO signal in Southern tropical $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ compared to Southern mid-latitudes and global average would be expected ~~in this scenario for several of these scenarios~~. This is because both biomass burning and wetland emissions show strong maxima in the Southern tropics and should be the dominant sources in this latitudinal band (Kirschke et al., 2013). The detrended $[\text{CH}_4]$ records from SMO show such a signal, but one that explains only one third of the IAV and doesn't seem to have significant impact on the trends. Further, we don't find higher ENSO forcing of the $\delta^{13}\text{CH}_4$ variability even in the core region of its climatic impact. Corbett et al. (2017) show that during La Niña events high surface temperatures over the Western Pacific lead to upward transport over the Indonesian region (a CH_4 source area from wetlands and rice paddies) and negative CH_4 anomalies in the mid-troposphere (tropical surface air with relatively low $[\text{CH}_4]$ replaces air from the Northern Hemisphere with higher $[\text{CH}_4]$). This mechanism would dampen the signal of higher La Niña emissions in surface records like SMO and ASC. However, the corresponding El Niño anomalies in mid-tropospheric CH_4 over the Central Pacific are smaller. This indicates that Central Pacific surface air, where there are no CH_4 sources, is closer in $[\text{CH}_4]$ to mid-tropospheric levels than surface air from the Western Pacific. Unless there were strong longitudinal differences in mid-tropospheric $[\text{CH}_4]$, this is inconsistent with a scenario where high concentrations of CH_4 are generated over the Western Pacific in La Niñas but transported upwards and away from the surface stations used in this study. On hemispheric or global scales, transport processes are unlikely to play a strong role, given the short mixing time of methane relative to its atmospheric turn-over.

The low correlations of $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ with ENSO rule out a dominant role for ENSO triggered sink changes in atmospheric methane records. Removal processes could lead to either amplification or dampening of source signals. Higher emissions of methane and carbon monoxide from biomass burning will draw down OH and weaken the sink. Emission factors from fires for CO are between 10 and 30-fold higher than for CH_4 (Van der Werf et al., 2017), so that the biomass burning dynamics dominate the source of reactive carbon, leaving less OH during El Niños and more during La Niñas to draw down CH_4 . This would provide a negative feedback for the emissions $[\text{CH}_4]$ -signal from ENSO forcing. In contrast, the feedback on the ENSO emissions $\delta^{13}\text{CH}_4$ -signal would be positive due to varying enrichment of ^{13}C -methane through sink fractionation (less removal leads to less ^{13}C -enrichment of relatively ^{13}C -depleted wetland emissions during La Niñas; more removal increases the ^{13}C -enrichment from biomass burning emissions during El Niños further). In addition to the reactive carbon effect, Turner et al. (2018) found a further OH increase during La Niñas due to higher lightning rates with NO_x production. Turner et al. (2018) could attribute 17% of OH variability that is forced by climate cycles (rather than emissions of other atmospheric compounds) to ENSO. This is a minor part of the variability, but in consequence the dampening effect on $[\text{CH}_4]$ and the reinforcing feedback on $\delta^{13}\text{CH}_4$ would add further to the reactive carbon feedbacks. In our correlation results these sink impacts are not apparent, as the $[\text{CH}_4]$ correlations for the tropical stations are higher than $\delta^{13}\text{CH}_4$ correlations (Tables 2 and 3). Nevertheless, the OH-dynamics provide a possible explanation for the limited ENSO impact on $[\text{CH}_4]$ variability and trends. They also make $\delta^{13}\text{CH}_4$ a conservative proxy for the influence that ENSO exerts on tropical methane.

Whether ENSO has less influence on CH_4 emissions than assumed or whether such an impact is overwhelmed by atmospheric removal or other CH_4 cycle processes, our results suggest that global atmospheric trends in $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ are dominated by other components in the methane budget.

6. Conclusions

To study the impact of natural climate variability on recent trends in atmospheric methane concentration, we investigated the correlation between ENSO cycles and records of the mixing ratios/mole fractions and stable carbon isotopes of methane, as well as HCN as a biomass burning indicator. As $\delta^{13}\text{CH}_4$ is subject to a mutually reinforcing signal from ENSO suppression of wetland emissions and enhancement of biomass burning CH_4 (or vice versa), as well as positive feedbacks from OH-dynamics, it is particularly suited to study the role of ENSO in the CH_4 cycle.

We find a sizeable effect of ENSO on biomass burning, as indicated by HCN variability in Southern mid-latitudes. In contrast, ENSO explains a smaller fraction ($\leq 37\%$) of $\delta^{13}\text{CH}_4/[\text{CH}_4]$ IAV even in the Southern tropics, where the expected effect should be greatest. Trends in $[\text{CH}_4]$ and $\delta^{13}\text{CH}_4$ in these latitudes are far less influenced by ENSO ($\leq 23\%$). On hemispheric and global scales the ENSO signal in the methane records is similarly weak. Our results do not rule out that ENSO influences CH_4 emissions from wetlands and biomass burning through temperature, enhanced precipitation or droughts in key regions, but any such impacts are overwhelmed by OH-dynamics or other source and sink processes. We review literature estimates of ENSO-driven emissions and find them too small and sporadic to account for the post-2007 rise. Counteracting OH-dynamics are

~~expected to further dampen any influence ENSO may have on methane growth rates.~~ Our findings suggest that ENSO is not an important driver for recent global trends in methane, including the [CH₄] plateau and the increase in [CH₄] since 2007. The latter ~~rise~~ must therefore have different causes. Our results ~~cannot do not~~ rule out that wetland production is a ~~main~~ contributor to the post-2007 [CH₄]-rise if driven by environmental controls other than ENSO. ~~However, causes for a rapid shift in wetland production to match the sudden onset of the concentration rise in 2007 are unclear and a recent study does not find~~This is suggested by an increase in ~~global~~ wetland CH₄ production between the periods 2000-2006 and 2012 (Poulter 2006-2014, although with the limited confidence of a single wetland emissions model (Zhang et al., 2017)-2018). The longer the atmospheric [CH₄] and δ¹³CH₄ trends persist, the less probable are processes that impact IAV and short-lived cyclical events like ENSO as the driver. Therefore, we consider increasing anthropogenic sources as the more likely cause of the [CH₄]-rise. Changes in removal rates via OH have been suggested as an additional (Rigby et al., 2017) or alternative (Turner et al., 2017) driver of the increase, but recent work suggests that sink impacts are not dominant (Naus et al., 2018). There is evidence for additional methane emissions from agriculture (~~Schaefer (Wolf et al., 2016) 2017~~) and from fossil fuel sources (Hausmann et al., 2016) ~~and~~; both may contribute to the current rise in [CH₄] (Worden et al., 2017). Further identification of these ~~emissions processes~~ is necessary to inform climate change mitigation policies and climate projections.

15

7. Team list

Hinrich Schaefer¹, Dan Smale¹, Sylvia E. Nichol¹, Tony M. Bromley¹, Gordon W. Brailsford¹, Ross J. Martin¹, Rowena Moss¹, Sylvia Englund Michel², James W. C. White²

¹Climate and Atmosphere Centre, National Institute of Water and Atmospheric Research, Wellington, 6021, New Zealand

20 ² Institute of Arctic and Alpine Research, Boulder, CO, USA.

8. Copyright statement

Code availability: Not applicable

Data availability

25 Raw data for individual stations measured by INSTAAR and NIWA are available from the World Data Centre for Greenhouse Gases <http://ds.data.jma.go.jp/gmd/wdcgg/introduction.html>; [CH₄] and δ¹³CH₄ data from NOAA_ESRL and INSTAAR are also available from ftp://afp.cmdl.noaa.gov/data/trace_gases. Data from Heidelberg University are available from <http://www.iup.uni-heidelberg.de/institut/forschung/groups/kk/en/Data.html>.

~~Global δ¹³CH₄ data products are available on request from the corresponding author.~~

30 Lauder MIR-FTIR HCN is publicly available from the NDACC archive:

<ftp.cpc.ncep.noaa.gov/ndacc/station/lauder/hdf/ftir>

All data products and time series used in this study are available on request from the corresponding author.

35 Appendices

Supplement link (will be included by Copernicus)

Author contribution: H.S. designed the study, conducted the correlation analysis and prepared the manuscript. ~~E.D. supplied [CH₄] data.~~ T.M.B., R.J.M., R.M., G.W.B., S.-E.M., and J.W.C.W. supplied $\delta^{13}\text{C}$ data. D.S. supplied HCN data. S.E.N. conducted data processing and analysis.

5

14. Acknowledgements

We thank B. Graham, E. Behrens, Brett Mullan and S. Mikaloff Fletcher for helpful discussions and advice. Ed Dlugokencky from NOAA ESRL supplied [CH₄] data. D. Lowe started the BHD measurements that made this analysis possible. This project was supported by NIWA funding under Climate and Atmosphere Research Programme CAAC1804 (2017/18 SCI).

10

15. Disclaimer

16. References

- 15 Ashok, K., Behera, S.K., Rao, S.A., Weng, H.Y. and Yamagata, T.: El Nino Modoki and its possible teleconnection, *J. Geophys. Res.*, 112, 2007.
- Barnston, A.G., Chelliah, M. and Goldenberg, S.B.: Documentation of a highly ENSO-related SST region in the equatorial Pacific, *Atmos. Ocean*, 35, 367-383, 1997.
- 20 Basso, L.S., Gatti, L.V., Gloor, M., Miller, J.B., Domingues, L.G., Correia, C.S.C. and Borges, V.F.: Seasonality and interannual variability of CH₄ fluxes from the eastern Amazon Basin inferred from atmospheric mole fraction profiles, *J. Geophys. Res.*, 121, 168-184, 2016.
- 25 Bousquet, P., Ciais, P., Miller, J.B., Dlugokencky, E.J., Hauglustaine, D.A., Prigent, C., Van der Werf, G.R., Peylin, P., Brunke, E.-G., Carouge, C., Langenfelds, R.L., Lathiere, J., Papa, F., Ramonet, M., Schmidt, M., Steele, L.P., Tyler, S.C. and White, J.: Contribution of anthropogenic and natural sources to atmospheric methane variability, *Nature*, 443, doi:10.1038, 2006.
- 30 Bousquet, P., Ringeval, B., Pison, I., Dlugokencky, E.J., Brunke, E.G., Carouge, C., Chevallier, F., Fortems-Cheiney, A., Frankenberg, C., Hauglustaine, D.A., Krummel, P.B., Langenfelds, R.L., Ramonet, M., Schmidt, M., Steele, L.P., Szopa, S., Yver, C., Viovy, N. and Ciais, P.: Source attribution of the changes in atmospheric methane for 2006-2008, *Atmos. Chem. Phys.*, 11, 3689-3700, 2011.
- 35 Bridgman, S.D., Cadillo-Quiroz, H., Keller, J.K. and Zhuang, Q.L.: Methane emissions from wetlands: biogeochemical, microbial, and modeling perspectives from local to global scales, *Global Change Biol.*, 19, 1325-1346, 2013.
- 40 Chamberlain, S.D., Gomez-Casanovas, N., Walter, M.T., Boughton, E.H., Bernacchi, C.J., DeLucia, E.H., Groffman, P.M., Keel, E.W. and Sparks, J.P.: Influence of transient flooding on methane fluxes from subtropical pastures, *J. Geophys. Res.*, 121, 965-977, 2016.

- Cleveland, R.B., Cleveland, W.S., McRae, J.E. and Terpenning, I.: STL: A seasonal-trend decomposition procedure based on Loess, *J. Off. Stat.*, 6, 1990.
- 5 [Corbett, A., Jiang, X., Xiong, X.Z., Kao, A. and Li, L.M.: Modulation of midtropospheric methane by El Nino. *Earth Space Sci.*, 4, 590-596, 2017.](#)
- Curtis, S. and Adler, R.: ENSO indices based on patterns of satellite-derived precipitation, *J. Climate*, 13, 2786-2793, 2000.
- 10 Dlugokencky, E.J., Lang, P.M., Crotwell, A.M., Mund, J.W., Crotwell, M.J. and Thoning, K.W.: Atmospheric Methane Dry Air Mole Fractions from the NOAA ESRL Carbon Cycle Cooperative Global Air Sampling Network, 1983-2016, in: Division, N.-E.G.M. (Ed.), Version: 2017-07-28, , Path: ftp://aftp.cmdl.noaa.gov/data/trace_gases/ch4/flask/surface/, 2017
- 15 Dlugokencky, E.J., [Myers, R.C., Lang, P.M., Masarie, K.A., Crotwell, A.M., Thoning, K.W., Hall, B.D., Elkins, J.W. and Steele, L.P.: Conversion of NOAA atmospheric dry air CH4 mole fractions to a gravimetrically prepared standard scale. *J. Geophys. Res.*, 110, doi:10.1029/2005JD006035, 2005.](#)
- [Dlugokencky, E.J.](#), Nisbet, E.G., Fisher, R. and Lowry, D.: Global atmospheric methane: budget, changes and dangers, *Phil. Trans. Royal Soc. A*, 369, 2058-2072, 2011.
- 20 Ferretti, D.F., Miller, J.B., White, J.W.C., Etheridge, D.M., Lassey, K.R., Lowe, D.C., MacFarling Meure, C.M., Dreier, M.F., Trudinger, C.M., van Ommen, T.D. and Langenfelds, R.L.: Unexpected changes to the global methane budget over the past 2000 years, *Science*, 309, 1714-1717, 2005.
- 25 Hausmann, P., Sussmann, R. and Smale, D.: Contribution of oil and natural gas production to renewed increase in atmospheric methane (2007-2014): top-down estimate from ethane and methane column observations, *Atmos. Chem. Phys.*, 16, 3227-3244, 2016.
- 30 Hodson, E.L., Poulter, B., Zimmermann, N.E., Prigent, C. and Kaplan, J.O.: The El Nino-Southern Oscillation and wetland methane interannual variability, *Geophys. Res. Lett.*, 38, 2011.
- Holmes, M.E., Chanton, J.P., Tfaily, M.M. and Ogram, A.: CO₂ and CH₄ isotope compositions and production pathways in a tropical peatland, *Global Biogeochem. Cy.*, 29, 1-18, 2015.
- 35 Horel, J.D. and Wallace, J.M.: Planetary-Scale Atmospheric Phenomena Associated with the Southern Oscillation, *Mon. Weather Rev.*, 109, 813-829, 1981.
- 40 Houweling, S., Krol, M., Bergamaschi, P., Frankenberg, C., Dlugokencky, E.J., Morino, I., Notholt, J., Sherlock, V., Wunch, D., Beck, V., Gerbig, C., Chen, H., Kort, E.A., Rockmann, T. and Aben, I.: A multi-year methane inversion using SCIAMACHY, accounting for systematic errors using TCCON measurements, *Atmos. Chem. Phys.*, 14, 3991-4012, 2014.
- 45 Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J.G., Dlugokencky, E.J., Bergamaschi, P., Bergmann, D., Blake, D.R., Bruhwiler, L., Cameron-Smith, P., Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E.L., Houweling, S., Josse, B., Fraser, P.J., Krummel, P.B., Lamarque, J.F., Langenfelds, R.L., Le Quere, C., Naik, V., O'Doherty, S., Palmer, P.I., Pison, I., Plummer, D., Poulter, B., Prinn, R.G., Rigby, M., Ringeval, B., Santini, M., Schmidt, M., Shindell, D.T., Simpson, I.J., Spahni, R., Steele, L.P., Strode, S.A., Sudo, K., Szopa, S., van der Werf, G.R., Voulgarakis, A., van Weele, M., Weiss, R.F., Williams, J.E. and Zeng, G.: Three decades of global methane sources and sinks, *Nat. Geosci.*, 6, 813-823, 2013.

- Kousky, V.E. and Higgins, R.W.: An alert classification system for monitoring and assessing the ENSO cycle, *Weather Forecast*, 22, 353-371, 2007.
- 5 Li, Q.B., Jacob, D.J., Bey, I., Yantosca, R.M., Zhao, Y.J., Kondo, Y. and Notholt, J.: Atmospheric hydrogen cyanide (HCN): Biomass burning source, ocean sink? *Geophys. Res. Lett.*, 27, 357-360, 2000.
- Liu, J.J., Bowman, K.W., Schimel, D.S., Parazoo, N.C., Jiang, Z., Lee, M., Bloom, A.A., Wunch, D., Frankenberg, C., Sun, Y., O'Dell, C.W., Gurney, K.R., Menemenlis, D., Gierach, M., Crisp, D. and Eldering, A.: Contrasting carbon cycle responses of the tropical continents to the 2015-2016 El Nino, *Science*, 358, 191-193, 2017.
- 10 [Lowe, D.C., Brenninkmeijer, C.A.M., Tyler, S.C., et al.: Determination of the isotopic composition of atmospheric methane and its application in the Antarctic, *J. Geophys. Res.*, 96, 15455-15467, 1991.](#)
- 15 Lyon, B. and Barnston, A.G.: ENSO and the spatial extent of interannual precipitation extremes in tropical land areas, *J. Climate*, 18, 5095-5109, 2005.
- MacFarling Meure, C., Etheridge, D., Trudinger, C., Steele, P., Langenfelds, R., van Ommen, T., Smith, A. and Elkins, J.: Law Dome CO₂, CH₄ and N₂O ice core records extended to 2000 years BP, *Geophys. Res. Lett.*, 33, 2006.
- 20 [McNorton, J., Chipperfield, M.P., Gloor, M., Wilson, C., Feng, W.H., Hayman, G.D., Rigby, M., Krummel, P.B., O'Doherty, S., Prinn, R.G., Weiss, R.F., Young, D., Dlugokencky, E. and Montzka, S.A.: Role of OH variability in the stalling of the global atmospheric CH₄ growth rate from 1999 to 2006, *Atmos. Chem. Phys.*, 16, 7943-7956, 2016.](#)
- 25 McPhaden, M.J., Zebiak, S.E. and Glantz, M.H.: ENSO as an integrating concept in Earth science, *Science*, 314, 1740-1745, 2006.
- Melton, J.R., Wania, R., Hodson, E.L., Poulter, B., Ringeval, B., Spahni, R., Bohn, T., Avis, C.A., Beerling, D.J., Chen, G., Eliseev, A.V., Denisov, S.N., Hopcroft, P.O., Lettenmaier, D.P., Riley, W.J., Singarayer, J.S., Subin, Z.M., Tian, H., Zurcher, S., Brovkin, V., van Bodegom, P.M., Kleinen, T., Yu, Z.C. and Kaplan, J.O.: Present state of global wetland extent and wetland methane modelling: conclusions from a model inter-comparison project (WETCHIMP), *Biogeosciences*, 10, 753-788, 2013.
- 30 [Möller, L., Sowers, T., Bock, M., Spahni, R., Behrens, M., Schmitt, J., Miller, H., Fischer, H.: Independent variations of CH₄ emissions and isotopic composition over the past 160,000 years, *Nat. Geosci.* 6, 885-890, 2013.](#)
- 35 [Naus, S., Montzka, S.A., Pandey, S., Basu, S., Dlugokencky, E.J. and Krol, M.: Constraints and biases in a tropospheric two-box model of OH, *Atmos. Chem. Phys. Disc.*, 2018.](#)
- 40 NDACC, Infrared Working Group: IRWG Uniform Retrieval Parameter Summary, http://www.acom.ucar.edu/irwg/IRWG_Uniform_RP_Summary-3.pdf, 2014
- 45 Nisbet, E.G., Dlugokencky, E.J., Manning, M.R., Lowry, D., Fisher, R.E., France, J.L., Michel, S.E., Miller, J.B., White, J.W.C., Vaughn, B., Bousquet, P., Pyle, J.A., Warwick, N.J., Cain, M., Brownlow, R., Zazzeri, G., Lanoiselle, M., Manning, A.C., Gloor, E., Worthy, D.E.J., Brunke, E.G., Labuschagne, C., Wolff, E.W. and Ganesan, A.L.: Rising atmospheric methane: 2007-2014 growth and isotopic shift, *Global Biogeochem. Cy.*, 30, 1356-1370, 2016.
- Pandey, S., Houweling, S., Krol, M., Aben, I., Monteil, G., Nechita-Banda, N., Dlugokencky, E.J., Detmers, R., Hasekamp, O., Xu, X.Y., Riley, W.J., Poulter, B., Zhang, Z., McDonald, K.C., White, J.W.C., Bousquet, P. and Rockmann, T.: Enhanced methane emissions from tropical wetlands during the 2011 La Nina, *Sci. Rep.*, 2017.
- 50

- Parker, R.J., Boesch, H., McNorton, J., Comyn-Platt, E., Gloor, M., Wilson, C., Chipperfield, M.P., Hayman, G.D. and Bloom, A.A.: Evaluating year-to-year anomalies in tropical wetland methane emissions using satellite CH₄ observations, *Remote Sens. Environ.*, 211, 261-275, 2018.
- 5 Poulter, B., Bousquet, P., Canadell, J.G., Ciais, P., Peregon, A., Saunio, M., Arora, V.K., Beerling, D.J., Brovkin, V., Jones, C.D., Joos, F., Gedney, N., Ito, A., Kleinen, T., Koven, C.D., McDonald, K., Melton, J.R., Peng, C.H., Peng, S.S., Prigent, C., Schroeder, R., Riley, W.J., Saito, M., Spahni, R., Tian, H.Q., Taylor, L., Viovy, N., Wilton, D., Wiltshire, A., Xu, X.Y., Zhang, B.W., Zhang, Z. and Zhu, Q.A.: Global wetland contribution to 2000-2012 atmospheric methane growth rate dynamics, *Environ. Res. Lett.*, 12, 2017.
- 10 Prather, M.J., Holmes, C.D. and Hsu, J.: Reactive greenhouse gas scenarios: Systematic exploration of uncertainties and the role of atmospheric chemistry, *Geophys. Res. Lett.*, 39, 2012.
- Pumphrey, H.C., Glatthor, N., Bernath, P.F., Boone, C.D., Hannigan, J.W., Ortega, I., Livesey, N.J. and Read, W.G.: MLS measurements of stratospheric hydrogen cyanide during the 2015-2016 El Nino event, *Atmos. Chem. Phys.*, 18, 691-703, 2018.
- 15 Rice, A.L., Butenhoff, C.L., Teama, D.G., Roger, F.H., Khalil, M.A.K. and Rasmussen, R.A.: Atmospheric methane isotopic record favors fossil sources flat in 1980s and 1990s with recent increase, *Proc. Nat. Acad. Sci. USA*, 113, 10791-10796, 2016.
- 20 [Rigby, M., Montzka, S.A., Prinn, R.G., White, J.W.C., Young, D., O'Doherty, S., Lunt, M.F., Ganesan, A.L., Manning, A.J., Simmonds, P.G., Salameh, P.K., Harth, C.M., Muhle, J., Weiss, R.F., Fraser, P.J., Steele, L.P., Krummel, P.B., McCulloch, A. and Park, S.: Role of atmospheric oxidation in recent methane growth, *Proc. Nat. Acad. Sci. USA*, 114, 5373-5377, 2017.](#)
- 25 Ropelewski, C.F. and Halpert, M.S.: Global and Regional Scale Precipitation Patterns Associated with the El-Nino Southern Oscillation, *Mon. Weather Rev.*, 115, 1606-1626, 1987.
- Saunio, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J.G., Dlugokencky, E.J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F.N., Castaldi, S., Jackson, R.B., Alexe, M., Arora, V.K., Beerling, D.J., Bergamaschi, P., Blake, D.R., Brailsford, G., Brovkin, V., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry, C., Frankenberg, C., Gedney, N., Hoglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H.S., Kleinen, T., Krummel, P., Lamarque, J.F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K.C., Marshall, J., Melton, J.R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F.J.W., Patra, P.K., Peng, C.H., Peng, S.S., Peters, G.P., Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W.J., Saito, M., Santini, M., Schroeder, R., Simpson, I.J., Spahni, R., Steele, P., Takizawa, A., Thornton, B.F., Tian, H.Q., Tohjima, Y., Viovy, N., Voulgarakis, A., van Weele, M., van der Werf, G.R., Weiss, R., Wiedinmyer, C., Wilton, D.J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X.Y., Yoshida, Y., Zhang, B., Zhang, Z. and Zhu, Q.: The global methane budget 2000-2012, *Earth Syst. Sci. Data*, 8, 697-751, 2016.
- 30 Saunio, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J.G., Dlugokencky, E.J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F.N., Castaldi, S., Jackson, R.B., Alexe, M., Arora, V.K., Beerling, D.J., Bergamaschi, P., Blake, D.R., Brailsford, G., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Frankenberg, C., Gedney, N., Hoglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H.S., Kleinen, T., Krummel, P., Lamarque, J.F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., Melton, J.R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F.J., Patra, P.K., Peng, C.H., Peng, S.S., Peters, G.P., Pison, I., Prinn, R., Ramonet, M., Riley, W.J., Saito, M., Santini, M., Schroeder, R., Simpson, I.J., Spahni, R., Takizawa, A., Thornton, B.F., Tian, H.Q., Tohjima, Y., Viovy, N., Voulgarakis, A., Weiss, R., Wilton, D.J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X.Y., Yoshida, Y., Zhang, B.W., Zhang, Z. and Zhu, Q.A.: Variability and quasi-decadal changes in the methane budget over the period 2000-2012, *Atmos. Chem. Phys.*, 17, 11135-11161, 2017.
- 40
- 45

- Schaefer, H., Fletcher, S.E.M., Veidt, C., Lassey, K.R., Brailsford, G.W., Bromley, T.M., Dlugokencky, E.J., Michel, S.E., Miller, J.B., Levin, I., Lowe, D.C., Martin, R.J., Vaughn, B.H. and White, J.W.C.: A 21st-century shift from fossil-fuel to biogenic methane emissions indicated by (CH₄)-C-13, *Science*, 352, 80-84, 2016.
- 5 Schwietzke, S., Sherwood, O.A., Ruhwiler, L.M.P.B., Miller, J.B., Etiope, G., Dlugokencky, E.J., Michel, S.E., Arling, V.A., Vaughn, B.H., White, J.W.C. and Tans, P.P.: Upward revision of global fossil fuel methane emissions based on isotope database, *Nature*, 538, 88-91, 2016.
- 10 Sheese, P.E., Walker, K.A. and Boone, C.D.: A global enhancement of hydrogen cyanide in the lower stratosphere throughout 2016, *Geophys. Res. Lett.*, 2017.
- Shindell, D.T., Faluvegi, G., Koch, D.M., Schmidt, G.A., Unger, N. and Bauer, S.E.: Improved Attribution of Climate Forcing to Emissions, *Science*, 326, 716-718, 2009.
- 15 Sweeney, C., Dlugokencky, E., Miller, C.E., Wofsy, S., Karion, A., Dinardo, S., Chang, R.Y.W., Miller, J.B., Bruhwiler, L., Crotwell, A.M., Newberger, T., McKain, K., Stone, R.S., Wolter, S.E., Lang, P.E. and Tans, P.: No significant increase in long-term CH₄ emissions on North Slope of Alaska despite significant increase in air temperature, *Geophys. Res. Lett.*, 43, 6604-6611, 2016.
- 20 Trenberth, K.E.: Spatial and Temporal Variations of Southern Oscillation. *Q J Roy Meteor Soc* 102, 639-653.
Troup, A.J. (1965) Southern Oscillation, *Quat. J. Roy. Meteor. Soc.*, 91, 1976.
- Turetsky, M.R., Kotowska, A., Bubier, J., Dise, N.B., Crill, P., Hornibrook, E.R.C., Minkinen, K., Moore, T.R., Myers-Smith, I.H., Nykanen, H., Olefeldt, D., Rinne, J., Saarnio, S., Shurpali, N., Tuittila, E.S., Waddington, J.M., White, J.R., Wickland, K.P. and Wilking, M.: A synthesis of methane emissions from 71 northern, temperate, and subtropical wetlands, *Global Change Biol.*, 20, 2183-2197, 2014.
- 30 [Turner, A.J., Frankenberg, C., Wennberg, P.O. and Jacob, D.J.: Ambiguity in the causes for decadal trends in atmospheric methane and hydroxyl, *Proc. Nat. Acad. Sci. USA*, 114, 5367-5372, 2017.](#)
- [Turner, A.J., Fung, I., Naik, V., Horowitz, L.W. and Cohen, R.C.: Modulation of hydroxyl variability by ENSO in the absence of external forcing, *Proc. Nat. Acad. Sci. USA*, 115, 8931-8936, 2018.](#)
- 35 van Aardenne, J.A., Dentener, F.J., Olivier, J.G.J., Goldewijk, C.G.M.K. and Lelieveld, J.: A 1 degrees x 1 degrees resolution data set of historical anthropogenic trace gas emissions for the period 1890-1990, *Global Biogeochem. Cy.*, 15, 909-928, 2001.
- van der Werf, G.R., Randerson, J.T., Giglio, L., Collatz, G.J., Kasibhatla, P.S. and Arellano, A.F.: Interannual variability in global biomass burning emissions from 1997 to 2004, *Atmos. Chem. Phys.*, 6, 3423-3441, 2006.
- 40 van der Werf, G.R., Randerson, J.T., Giglio, L., Collatz, G.J., Mu, M., Kasibhatla, P.S., Morton, D.C., DeFries, R.S., Jin, Y. and van Leeuwen, T.T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997-2009), *Atmos. Chem. Phys.*, 10, 11707-11735, 2010.
- 45 [Van der Werf, G.R., Randerson, J.T., Giglio, L., Van Leeuwen, T.T., Chen, Y., Rogers, B.M., Mu, M., van Marle, M.J.E., Morton, D.C., Collatz, G.J., Yokelson, R.J. and Kasibhatla, P.S.: Global fire emissions estimates during 1997–2016, *Earth Syst. Sci. Data* 9, 697–720, 2017.](#)
- [Wolf, J., Asrar, G.R. and West, T.O. and Revised methane emissions factors and spatially distributed annual carbon fluxes for global livestock, *Carbon Balance Management* 12, 2017.](#)
- 50

- Wolter, K. and Timlin, M.S.: Monitoring ENSO in COADS with a seasonally adjusted principal component index, Proc. 17th Climate Diagnostics Workshop, 1993.
- 5 Wolter, K. and Timlin, M.S.: Measuring the strength of ENSO events: How does 1997/98 rank? *Weather*, 53, 315-324, 1998.
- Worden, J.R., Bloom, A.A., Pandey, S., Jiang, Z., Worden, H.M., Walker, T.W., Houweling, S. and Rockmann, T.: Reduced biomass burning emissions reconcile conflicting estimates of the post-2006 atmospheric methane budget, *Nat. Commun.*, 8, 2017.
- 10 Xie, F., Li, J.P., Tian, W.S., Zhang, J.K. and Sun, C.: The relative impacts of El Nino Modoki, canonical El Nino, and QBO on tropical ozone changes since the 1980s, *Environ. Res. Lett.*, 9, 2014.
- Yeh, S.W., Kug, J.S., Dewitte, B., Kwon, M.H., Kirtman, B.P. and Jin, F.F.: El Nino in a changing climate, *Nature*, 462, 511-513, 2009.
- 15 Zeng, G., Wood, S.W., Morgenstern, O., Jones, N.B., Robinson, J. and Smale, D.: Trends and variations in CO, C₂H₆, and HCN in the Southern Hemisphere point to the declining anthropogenic emissions of CO and C₂H₆, *Atmos. Chem. Phys.*, 12, 7543-7555, 2012.
- 20 [Zhang, Z., Zimmermann, N.E., Calle, L., Hurtt, G., Chatterjee, A. and Poulter, B.: Enhanced response of global wetland methane emissions to the 2015-2016 El Nino-Southern Oscillation event, *Environ. Res. Lett.*, 13, 2018.](#)
- Zhu, Q., Peng, C., Chen, H., Fang, X., Liu, J., Jiang, H., Yang, Y. and Yang, G.: Estimating global natural wetland methane emissions using process modelling: spatio-temporal patterns and contributions to atmospheric methane fluctuations, *Global Ecol. Biogeogr.*, 24, 959-972, 2015.
- 25 Zhu, Q., Peng, C., Ciais, P., Jiang, H., Liu, J., Bousquet, P., Li, S., Chang, J., Fang, X. and Zhou, X.: Inter-annual Variation in Methane Emissions from Tropical Wetlands Triggered by Repeated El Niño Southern Oscillation, *Global Change Biol.*, 2017.

30

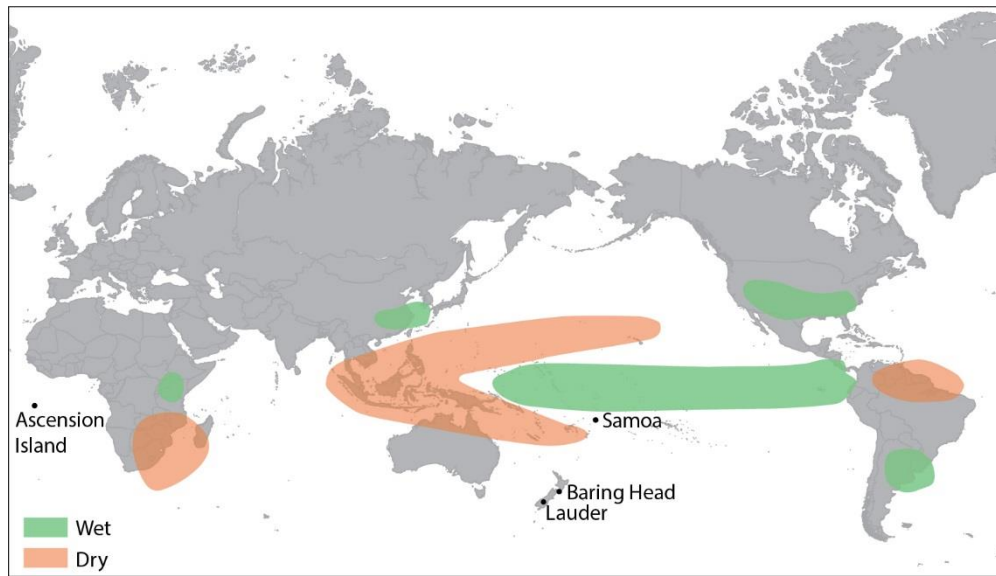


Fig. 1: Regions of ENSO impacts and monitoring stations used in this study.

The map indicates the locations of the atmospheric monitoring stations on Ascension Island (ASC), Samoa (SMO), Baring
 5 Head (BHD) and Lauder (LAU). General precipitation anomalies during northern hemisphere El Niño conditions for Dec-Feb
 are taken from <https://www.climate.gov/news-features/featured-images/global-impacts-el-nino-and-la-nina>. El Niño dry
 regions in Jun-Aug are similar for southern Asia and South America; during La Niña events opposite patterns for wet- and
 dryness develop in roughly the same regions.

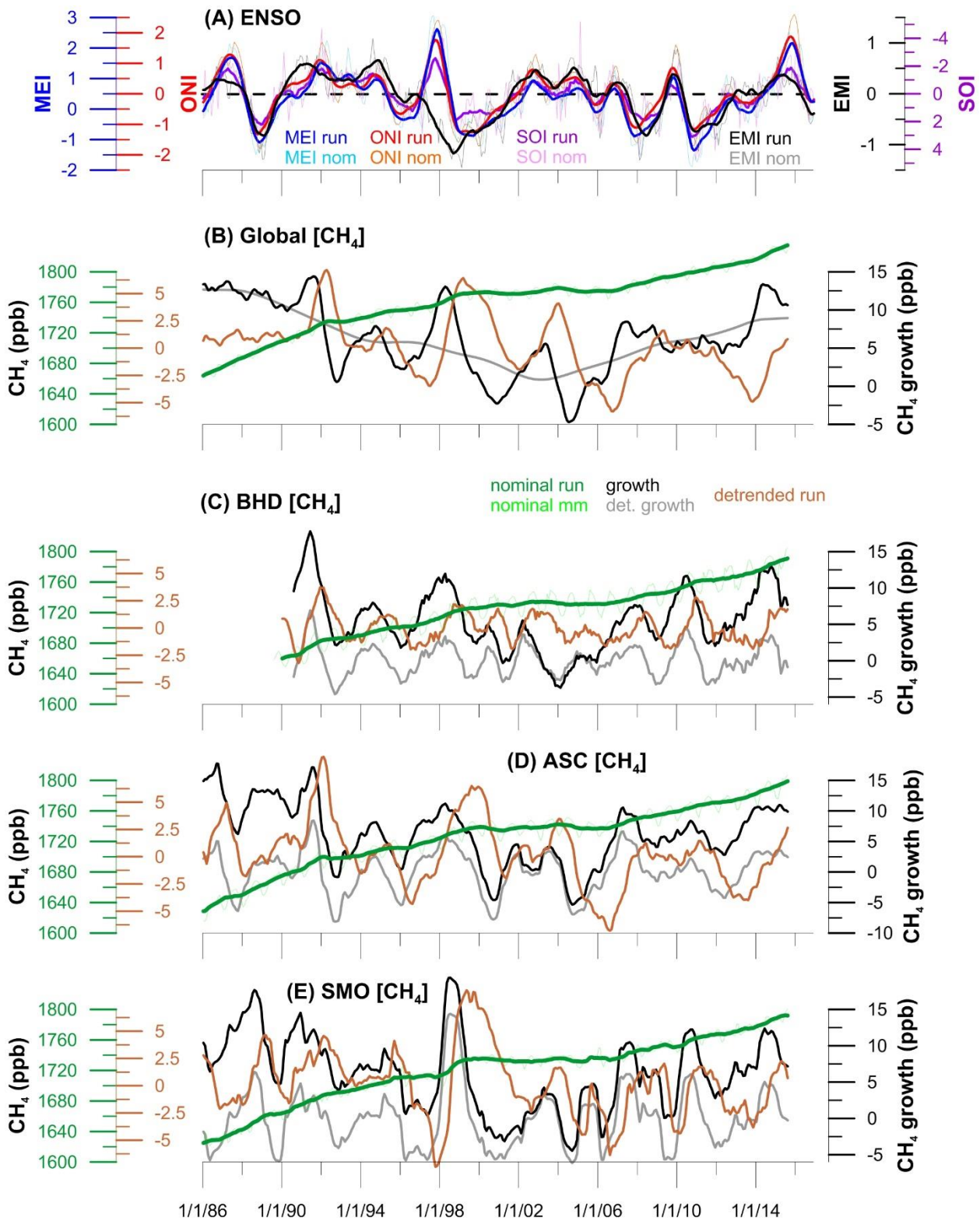


Fig. 2: Selected time series of ENSO indices and [CH₄]

Panels from top to bottom: (A) multivariate ENSO Index (MEI), Southern Oscillation Index (SOI), Ocean Niño Index (ONI), and El Modoki Index (EMI) shown for nominal literature data and their 12-month running means. (B) Global [CH₄] records; monthly means, 12-month running mean, detrended 12-month running mean, as well as nominal and detrended growth rates. (C) [CH₄] records from BHD (D) [CH₄] records from ASC. (E) [CH₄] records from SMO. BHD, ASC and SMO display same records as for global time series.

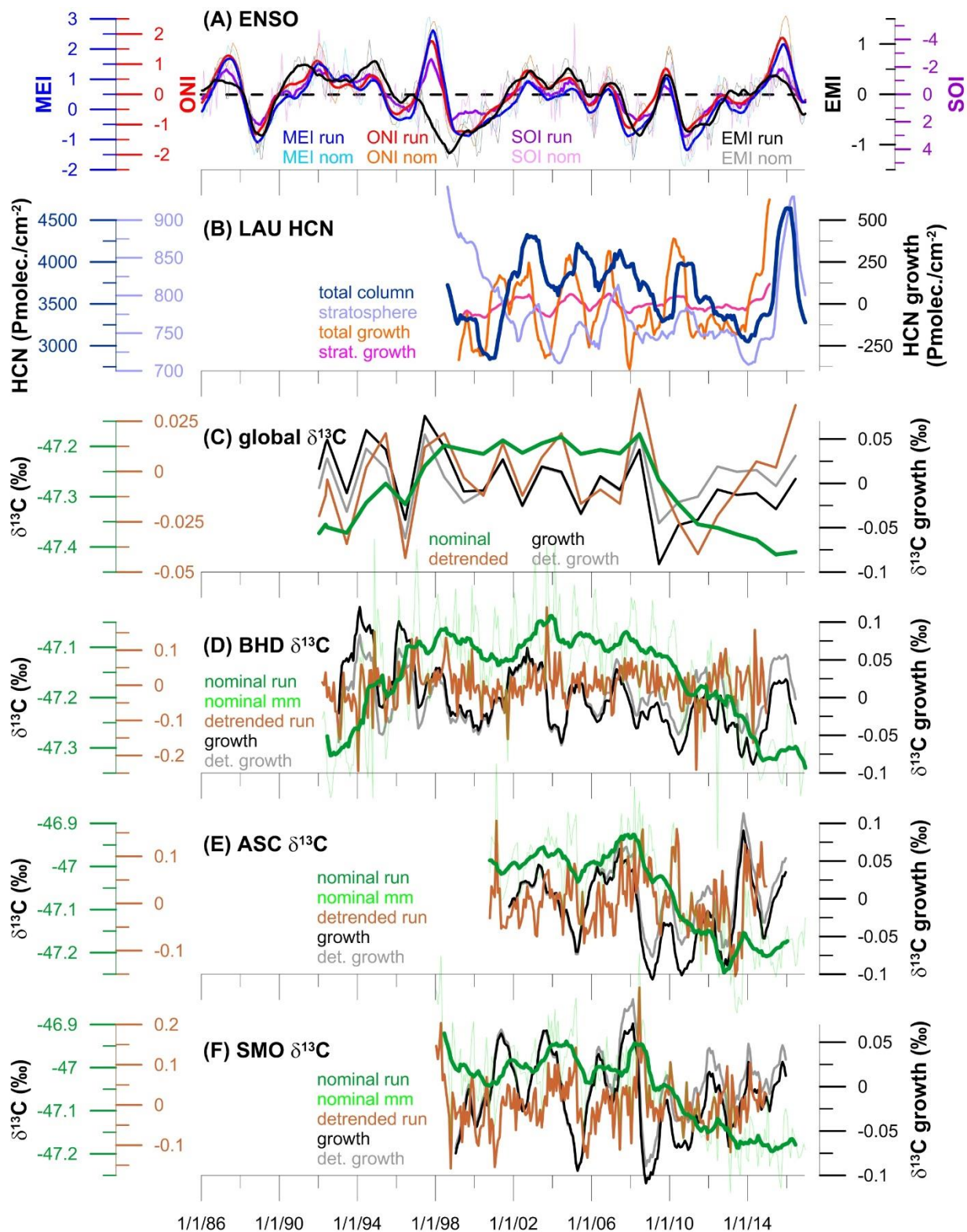


Fig. 3: Selected time series of ENSO indices, HCN and $\delta^{13}\text{CH}_4$

Panels from top to bottom: (A) multivariate ENSO Index (MEI), Southern Oscillation Index (SOI), Ocean Niño Index (ONI), and El Modoki Index (EMI) shown for nominal literature data and their 12-month running means. (B) HCN records as 12-month running means from LAU for total atmospheric column and stratosphere (12-100 km) and respective growth rates. (C) 5 Global annually averaged $\delta^{13}\text{CH}_4$ according to Schaefer et al. (2016) updated to end of 2016; nominal and detrended values and their respective growth rates. (D) $\delta^{13}\text{CH}_4$ from BHD; monthly means, 12-month running mean, detrended 12-month running mean, as well as nominal and detrended growth rates. (E) $\delta^{13}\text{CH}_4$ from ASC. (F) $\delta^{13}\text{CH}_4$ from SMO. ASC and SMO display same records as for BHD. Note scale differences between all $\delta^{13}\text{C}$ -axes to accentuate variability for comparison with ENSO.

| 10

Table 1: Description of time series products used in the correlation analyses.

<u>Parameter</u>	<u>Time series</u>	<u>Description</u>
<u>[CH₄]</u>	<u>global nom</u>	<u>global monthly means</u>
	<u>global gro</u>	<u>global monthly growth rates</u>
	<u>global run</u>	<u>12-month running mean of global monthly means</u>
	<u>global res</u>	<u>STL residual of global monthly means</u>
	<u>glob det-nom</u>	<u>detrended global monthly means</u>
	<u>glob det-gro</u>	<u>detrended global monthly growth rates</u>
	<u>glob det-run</u>	<u>detrended 12-month running mean of global monthly means</u>
	<u>δ¹³CH₄ and [CH₄]</u>	<u>NNN* nom</u>
<u>NNN* gro</u>		<u>station monthly growth rates</u>
<u>NNN* run</u>		<u>12-month running mean of station monthly means</u>
<u>NNN* res</u>		<u>STL residual of station monthly means</u>
<u>NNN* det-nom</u>		<u>detrended station monthly means</u>
<u>NNN* det-gro</u>		<u>detrended station monthly growth rates</u>
<u>NNN* det-run</u>		<u>detrended 12-month running mean of station monthly means</u>
<u>HCN (LAU)</u>		<u>total nom</u>
	<u>total gro</u>	<u>Total column monthly growth rates</u>
	<u>total run</u>	<u>Total column 12-month running mean of station monthly means</u>
	<u>total res</u>	<u>Total column STL residual of station monthly means</u>
	<u>strat. nom</u>	<u>Stratosphere monthly means</u>
	<u>strat. gro</u>	<u>Stratosphere monthly growth rates</u>
	<u>strat. run</u>	<u>Stratosphere 12-month running mean of station monthly means</u>
	<u>strat. res</u>	<u>Stratosphere STL residual of station monthly means</u>
<u>δ¹³CH₄</u>	<u>global nom</u>	<u>global yearly means</u>
	<u>global gro</u>	<u>global yearly growth rates</u>
	<u>global det</u>	<u>detrended global yearly means</u>
	<u>global det-gro</u>	<u>detrended global yearly growth rates</u>

*NNN as station acronym ASC, BHD, or SMO

Table 2: Spearman correlation of methane ~~mixing ratio~~ mole fraction with ENSO variability.

Correlations (r^2 -values) for the Spearman ranking coefficient between $[\text{CH}_4]$ time series from various sites and ENSO indices with lag times (in months) for optimum results. Colour backgrounds indicate r^2 -values in 10% classes. Grey background indicates correlations with p-values > 0.001.

5

Time series	MEI nom		MEI run		ONI nom		ONI run		SOI nom		SOI run		EMI nom		EMI run	
	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag
[CH ₄]																
global nom	0.08	59	0.12	58	0.04	48	0.06	50	0.10	59	0.17	54	0.04	5	0.03	10
global gro	0.10	6	0.10	6	0.08	7	0.09	59	0.06	8	0.08	33	0.06	50	0.09	52
global run	0.10	56	0.18	53	0.06	55	0.09	51	0.11	57	0.24	54	0.07	8	0.08	10
global res	0.06	49	0.06	47	0.09	49	0.11	48	0.04	25	0.06	60	0.10	0	0.09	0
glob det-nom	0.10	49	0.02	49	0.06	49	0.04	50	0.05	29	0.02	60	0.07	9	0.03	5
glob det-gro	0.02	58	0.04	60	0.00	58	0.00	59	0.02	59	0.06	60	0.03	0	0.04	0
glob det-run	0.05	49	0.06	48	0.09	48	0.12	49	0.03	0	0.04	60	0.08	0	0.08	1
BHD nom	0.10	56	0.11	58	0.05	45	0.05	47	0.11	56	0.15	59	0.07	53	0.05	51
BHD gro	0.08	24	0.10	27	0.10	51	0.11	55	0.08	24	0.12	24	0.15	60	0.19	60
BHD run	0.05	44	0.11	44	0.04	0	0.07	0	0.08	45	0.17	53	0.08	7	0.09	7
BHD res	0.03	46	0.02	16	0.02	16	0.02	16	0.02	16	0.02	14	0.02	33	0.02	29
BHD det-nom	0.07	44	0.01	17	0.02	33	0.01	36	0.03	13	0.01	17	0.04	30.00	0.00	29
BHD det-gro	0.13	56	0.17	58	0.12	23	0.13	59	0.10	23	0.14	59	0.08	60	0.08	24
BHD det-run	0.10	60	0.14	60	0.07	44	0.09	42	0.07	60	0.11	60	0.08	33	0.11	32
ASC nom	0.09	56	0.11	45	0.05	44	0.06	46	0.10	42	0.16	46	0.05	53	0.04	50
ASC gro	0.09	29	0.13	31	0.11	53	0.13	55	0.06	31	0.11	32	0.15	50	0.21	53
ASC run	0.08	43	0.16	45	0.07	44	0.10	44	0.11	43	0.22	43	0.06	46	0.06	47
ASC res	0.08	42	0.09	18	0.12	42	0.12	42	0.06	41	0.06	60	0.08	0	0.07	2
ASC det-nom	0.11	43	0.02	45	0.06	43	0.03	45	0.06	43	0.01	44	0.06	4	0.02	2
ASC det-gro	0.20	10	0.26	10	0.18	10	0.21	11	0.14	10	0.20	10	0.09	51	0.12	53
ASC det-run	0.09	40	0.14	17	0.13	41	0.15	42	0.05	17	0.08	17	0.09	0	0.08	1
SMO nom	0.07	56	0.12	58	0.03	45	0.04	48	0.08	56	0.16	59	0.02	10	0.02	50
SMO gro	0.19	17	0.18	10	0.15	10	0.17	11	0.10	9	0.12	10	0.13	46	0.17	49
SMO run	0.07	51	0.14	53	0.04	49	0.05	51	0.08	53	0.18	55	0.01	7	0.01	10
SMO res	0.16	0	0.13	1	0.22	1	0.15	1	0.14	0	0.15	2	0.17	0	0.14	3
SMO det-nom	0.15	0	0.11	1	0.18	0	0.14	1	0.09	0	0.12	2	0.10	0	0.11	3
SMO det-gro	0.31	9	0.36	10	0.31	10	0.35	11	0.22	9	0.33	11	0.10	10	0.11	12
SMO det-run	0.26	1	0.24	2	0.29	2	0.27	3	0.21	2	0.25	2	0.23	2	0.21	4

Table 23: Spearman correlation of $\delta^{13}\text{CH}_4$ and HCN with ENSO variability.

5 Correlations (r^2 -values) for the Spearman ranking coefficient between dependent variables, i.e. $\delta^{13}\text{CH}_4$ and HCN time series from various sites, and ENSO indices with lag times (in months) for optimum results. Colour backgrounds indicate r^2 -values in 10% classes. Grey background indicates correlations with p-values > 0.001.

Time series	MEI nom		MEI run		ONI nom		ONI run		SOI nom		SOI run		EMI nom		EMI run	
	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag	r^2	lag
HCN (LAU)																
total nom	0.13	5	0.03	6	0.06	5	0.05	8	0.08	4	0.04	8	0.10	14	0.05	12
total gro	0.21	1	0.26	2	0.27	1	0.27	2	0.18	0	0.30	1	0.19	0	0.27	1
total run	0.23	7	0.30	9	0.35	7	0.39	9	0.23	8	0.38	8	0.40	9	0.51	10
total res	0.10	7	0.09	7	0.13	7	0.13	7	0.10	10	0.13	6	0.08	9	0.10	10
strat. nom	0.10	10	0.05	11	0.05	41	0.03	0	0.08	40	0.05	43	0.09	39	0.08	0
strat. gro	0.36	0	0.43	0	0.40	0	0.44	1	0.22	0	0.37	0	0.21	0	0.30	0
strat. run	0.03	37	0.05	38	0.05	0	0.08	0	0.05	40	0.08	41	0.07	18	0.10	0
strat. res	0.10	7	0.07	7	0.10	7	0.08	8	0.07	8	0.07	8	0.11	44	0.10	38
$\delta^{13}\text{CH}_4$																
global nom	0.17	2	0.17	49	0.13	2	0.12	50	0.27	46	0.27	49	0.16	4	0.05	47
global gro	0.20	39	0.18	41	0.18	39	0.16	41	0.34	49	0.20	15	0.15	4	0.08	16
global det	0.14	1	0.19	14	0.08	20	0.12	16	0.37	22	0.27	17	0.13	15	0.12	16
global det-gro	0.23	58	0.16	58	0.18	59	0.18	60	0.37	55	0.24	56	0.15	15	0.12	16
BHD nom	0.09	56	0.09	60	0.04	56	0.04	59	0.09	55	0.13	60	0.07	42	0.06	39
BHD gro	0.05	2	0.06	2	0.06	2	0.07	3	0.08	2	0.13	2	0.10	44	0.11	44
BHD run	0.09	14	0.14	16	0.11	15	0.15	17	0.10	15	0.20	17	0.07	29	0.09	36
BHD res	0.03	6	0.03	10	0.04	7	0.03	9	0.04	6	0.03	10	0.09	33	0.09	36
BHD det-nom	0.07	8	0.01	8	0.03	7	0.02	9	0.05	6	0.01	10	0.06	42	0.05	38
BHD det-gro	0.07	1	0.09	1	0.09	1	0.11	2	0.09	2	0.13	12	0.10	45	0.11	45
BHD det-run	0.07	9	0.09	11	0.08	12	0.10	12	0.06	9	0.09	13	0.15	32	0.20	35
ASC nom	0.08	54	0.08	44	0.05	54	0.04	45	0.08	55	0.13	45	0.05	52	0.03	49
ASC gro	0.10	13	0.14	13	0.10	13	0.09	13	0.11	12	0.22	12	0.13	9	0.13	7
ASC run	0.08	22	0.13	23	0.08	22	0.10	22	0.12	22	0.23	22	0.12	20	0.14	20
ASC res	0.03	17	0.04	21	0.03	16	0.02	19	0.05	18	0.06	20	0.03	14	0.03	19
ASC det-nom	0.05	18	0.03	22	0.03	17	0.01	19	0.06	18	0.04	17	0.05	14	0.01	16

ASC det-gro	0.07	13	0.07	14	0.05	13	0.05	60	0.06	12	0.11	13	0.07	8	0.05	6
ASC det-run	0.04	22	0.05	60	0.04	60	0.04	60	0.07	22	0.09	22	0.06	32	0.06	32
SMO nom	0.06	56	0.08	55	0.04	43	0.04	44	0.08	56	0.12	42	0.04	40	0.05	38
SMO gro	0.02	13	0.03	24	0.02	23	0.03	25	0.04	0	0.06	13	0.10	43	0.10	44
SMO run	0.07	15	0.12	18	0.08	16	0.10	19	0.09	16	0.19	20	0.06	19	0.07	21
SMO res	0.09	0	0.08	0	0.10	1	0.08	2	0.09	1	0.07	3	0.06	30	0.06	35
SMO det-nom	0.06	0	0.06	1	0.08	1	0.06	1	0.06	3	0.05	3	0.06	30	0.05	35
SMO det-gro	0.05	23	0.06	24	0.04	23	0.06	25	0.03	0	0.03	0	0.09	42	0.08	42
SMO det-run	0.17	1	0.20	1	0.20	2	0.22	2	0.12	4	0.16	3	0.13	26	0.14	30

Table 34: Pearson correlation of $\delta^{13}\text{CH}_4$ and HCN with ENSO variability.

Correlations (r^2 -values) for the Pearson correlation coefficient between dependent variables, i.e. $\delta^{13}\text{CH}_4$ and HCN time series from various sites, and ENSO indices with lag times (in months) for optimum results. Colour backgrounds indicate r^2 -values in 10% classes. Results have not been screened for p-values.

5

Time series	MEI nom		MEI run		ONI nom		ONI run		SOI nom		SOI run		EMI nom		EMI run	
	r ²	lag	r ²	lag	r ²	lag	r ²	lag	r ²	lag	r ²	lag	r ²	lag	r ²	lag
HCN (LAU)																
total nom	0.10	5	0.03	5	0.04	4	0.04	6	0.06	4	0.04	6	0.10	14	0.06	11
total gro	0.22	0	0.30	2	0.27	1	0.30	1	0.16	0	0.28	2	0.19	2	0.24	1
total run	0.22	4	0.29	6	0.34	6	0.40	7	0.18	6	0.34	8	0.36	10	0.46	11
total res	0.11	7	0.13	5	0.14	5	0.16	6	0.09	7	0.13	7	0.09	10	0.08	11
strat. nom	0.18	6	0.13	9	0.08	7	0.08	9	0.07	5	0.09	9	0.09	39	0.11	0
strat. gro	0.42	0	0.54	0	0.49	0	0.55	0	0.22	0	0.39	0	0.18	1	0.25	1
strat. run	0.12	14	0.17	12	0.05	11	0.07	13	0.05	17	0.11	13	0.18	0	0.18	0
strat. res	0.17	6	0.16	6	0.17	6	0.17	7	0.08	5	0.12	8	0.10	39	0.09	37
$\delta^{13}\text{CH}_4$																
global nom	0.16	1	0.16	50	0.14	2	0.09	50	0.24	46	0.25	39	0.13	3	0.05	50
global gro	0.20	39	0.14	42	0.16	39	0.13	43	0.28	49	0.20	15	0.12	41	0.10	0
global det	0.18	11	0.15	13	0.15	11	0.12	13	0.32	12	0.24	15	0.11	15	0.08	17
glob det-gro	0.14	40	0.13	58	0.13	58	0.12	59	0.28	49	0.14	59	0.16	15	0.10	16
BHD nom	0.10	56	0.09	60	0.04	0	0.05	0	0.10	55	0.14	60	0.05	42	0.04	39
BHD gro	0.07	61	0.10	60	0.08	0	0.09	1	0.08	2	0.14	1	0.14	45	0.17	47
BHD run	0.10	0	0.13	0	0.11	0	0.13	0	0.09	62	0.18	57	0.04	38	0.06	41
BHD res	0.05	6	0.05	9	0.06	7	0.05	9	0.06	6	0.04	10	0.07	35	0.05	9
BHD det-nom	0.07	8	0.02	7	0.03	7	0.02	8	0.06	6	0.02	9	0.05	42	0.04	34
BHD det-gro	0.10	0	0.13	0	0.13	0	0.15	1	0.07	2	0.13	0	0.12	45	0.14	47
BHD det-run	0.10	9	0.13	10	0.11	10	0.14	12	0.06	9	0.10	12	0.18	33	0.23	34
ASC nom	0.09	54	0.08	58	0.04	54	0.04	37	0.09	55	0.16	33	0.04	52	0.03	32
ASC gro	0.10	14	0.14	14	0.08	13	0.10	13	0.08	12	0.19	13	0.15	5	0.22	7
ASC run	0.07	51	0.11	54	0.04	50	0.06	53	0.09	51	0.20	27	0.05	22	0.08	23
ASC res	0.03	17	0.04	22	0.03	18	0.02	19	0.05	29	0.07	24	0.04	14	0.03	18
ASC det-nom	0.06	18	0.04	22	0.03	18	0.01	18	0.06	29	0.05	32	0.05	15	0.02	17
ASC det-gro	0.07	14	0.08	14	0.04	13	0.05	59	0.05	13	0.09	14	0.07	8	0.10	7
ASC det-run	0.07	22	0.09	23	0.04	59	0.05	59	0.07	25	0.16	24	0.06	56	0.08	57
SMO nom	0.07	56	0.08	54	0.03	45	0.04	45	0.09	42	0.14	45	0.05	41	0.05	39
SMO gro	0.04	66	0.04	67	0.04	66	0.03	25	0.03	0	0.06	0	0.10	43	0.11	43
SMO run	0.07	50	0.10	49	0.03	38	0.05	0	0.09	51	0.18	49	0.03	38	0.04	35
SMO res	0.10	0	0.07	2	0.10	1	0.07	3	0.10	1	0.06	5	0.06	14	0.06	16
SMO det-nom	0.07	0	0.06	2	0.08	1	0.05	3	0.07	1	0.05	5	0.06	14	0.05	16
SMO det-gro	0.05	22	0.05	24	0.05	66	0.06	25	0.02	26	0.03	26	0.06	42	0.07	42
SMO det-run	0.06	1	0.06	2	0.07	2	0.08	3	0.05	15	0.07	16	0.14	16	0.19	20

