# Response to Reviewers – Fest et al., 2016 bg-2016-181

Reviewer comments appear as normal text

Our responses appear in bold and italicised

#### **Reviewer 1**

Fest et al. aimed to understand CH4 dynamics in two Eucalyptus forests in Australia with different precipitation regimes. Fluxes of CH4 were measured in a high temporal resolution with six replications at each site. In addition, soil temperature and moisture, and inorganic N levels were measured. The data were analyzed using linear regression for casual correlation to explore which factors controlled CH4 dynamics. Fest et al. concluded that soil moisture regime could explain over 90% of the variability of CH4 dynamics.

I believe the strengths of this study is 1) very high temporal resolution in CH4 measurements and 2) air-filled porosity explained CH4 dynamics in almost the same manner for the two study sites. These are novel, and deserve publication.

We wish to thank the reviewer for a very thorough and constructive review. We agree with most of the suggestions and believe they greatly enhanced the quality of the paper.

However, the current manuscript is no more than a draft. The weaknesses includes 1) statistical analyses, 2) discussion are underdeveloped, and 3) it's not well written.

1) The data should be analyzed using a maximum likelihood framework with AIC or BIC to compare regressions and determine the importance of temperature.

We agree with the reviewer and have reanalysed the data as suggested by the reviewer: We used a maximum likelihood framework to arrive at the AICs for 3 different models (one model containing only soil temperature, one model containing only a measure of soil moisture (we choose AFP) and one model containing soil temperature and AFP as a predictor. The results of this analysis are displayed in Table R1:

Table R1: parameters and coefficients of determination (Adj. R²) of selected linear models in combination with results of a restricted maximum likelihood analysis (REML) explaining seasonal variability in mean chamber cycle methane flux (FCH4) at a mixed *Eucalyptus obliqua* forest stand, Wombat State Forest, Victoria (AU-WOM) and at a mixed *E. obliqua* and *E. regnans* forest stand, Warra LTER between, Tasmania, Australia (AU-WRR). Predictors: T<sub>S</sub> (soil temperature) and AFP (air-filled porosity). REML results: Akaike information criterion (AIC); Estimate of importance for models containing both predictors (*in parentheses*).

Site						
	Dependent	Constant	AFP	$T_{s}$		
	Variable	(Intercept)	(slope)	(slope)	AIC	Adj. R²
AU-WRR	F <sub>CH4</sub>	53.640	-195.378	-	5666	0.855
	F <sub>CH4</sub>	-19.543	-	-2.215	9657	0.158
	F <sub>CH4</sub>	55.587	-193.284 ( <b>0.997</b> )	-0.254 ( <b>0.003</b> )	5629	0.857
AU-WOM	F <sub>CH4</sub>	53.943	-195.768	-	7648	0.915
	F <sub>CH4</sub>	-6.320	-	-1.701	13088	0.209
	F <sub>CH4</sub>	54.766	-201.671 ( <b>0.998</b> )	0.147 ( <b>0.002</b> )	7617	0.900

The REML and AIC results confirm the interpretation of the original linear regression approach showing that soil moisture (in this case expressed as AFP) is the strongest predictor of soil CH<sub>4</sub> flux in both forest systems. The analysis also shows that the models including soil moisture and soil temperature perform marginally better based on AIC compared to models including only soil moisture to predict soil CH<sub>4</sub> flux. However, the importance rating of the predictors (soil moisture and soil temperature) clearly indicates that in both forest systems soil moisture dominates accounting more than 99% of the proportion of variance explained by the model compared to <0.01% proportion of the variance explained by soil temperature. This reconfirms our initial assessment of the datasets where we stated that including temperature as a variable improved the correlation with methane uptake to a small degree, but it did not improve the predictive capacity. However, it will be important for readers to understand that and we propose to include the AIC statistics in the revised manuscript.

We therefore have added the table above as new Table 2 to the manuscript and added following section to the methods:

"We used a restricted maximum likelihood framework (REML, automatic linear modelling in SPSS) to arrive at the Akaike information criterion for three selected models that predict soil CH<sub>4</sub> uptake (one model containing only soil temperature, one model containing only a measure of soil moisture (we choose AFP) and one model containing soil temperature and AFP as a predictors of soil CH<sub>4</sub> flux)."

We also added following section to the results:

"The AIC results of the REML analysis confirm the results of the linear regression approach (Table 2) showing that soil moisture (in this case expressed as AFP) is the strongest predictor of soil CH<sub>4</sub> flux in both forest systems. The analysis shows that the models including soil moisture and soil temperature perform marginally better based on AIC compared to models including only soil moisture to predict soil CH<sub>4</sub> flux. However, the importance rating of the predictors (soil moisture and soil temperature) clearly indicates that in both forest systems soil moisture dominates, as it accounts for more than 99% of the proportion of variance explained by the model compared to <0.01% proportion of the variance explained by soil temperature."

2) Discussion should emphasize the novelty of this study.

We have revised and restructured the discussion to emphasize the novelty of the study:

# The discussion now reads:

"One of the most novel results of our study is the strong linear relationship observed between soil moisture and CH<sub>4</sub> uptake. To our knowledge the strength of this relationship is unique for temperate forest systems measured using continuous automated chamber systems over a long-period. It is also striking that this strong linear relationship was similar in the two temperate eucalypt forests (dry and wet) regardless of the differences in forest structure, soil type, annual precipitation and geographical distance. It is possible that the two different measurement systems (GC at AU-WRR and FTIR at AU-WOM) could produce different measures of CH<sub>4</sub> flux if operated at the same site because of technological and methodological differences. If that were true, there would only be a remote chance that the two linear relationships between CH<sub>4</sub> flux and AFP would overlap one another. As such, our finding that the relationships between CH<sub>4</sub> flux and AFP do converge into one common regression line (as shown in Fig. 4) is worthy of note and suggests similar accuracy between the two measurement systems and similar function in soil CH<sub>4</sub> exchange processes at the two forest sites.

 $CH_4$  flux data collected long-term in temperate deciduous forest systems in Europe (Butterbach-Bahl and Papen, 2002) has shown that soil moisture can explain up to 58% of the seasonality in soil  $CH_4$  uptake. Similarly, Kiese et al. (2003) reported that soil moisture could explain up to 53% of the seasonality in  $CH_4$  exchange in a tropical rainforests in Queensland, Australia. Soil moisture influences soil gas diffusivity and is considered the most important factor controlling seasonality of  $CH_4$  uptake in soils worldwide (Dalal et al., 2008;Dalal and Allen, 2008;Smith et al., 2003;Smith et

al., 2000;Ball et al., 1997a) and the negative relationship between soil moisture and soil CH<sub>4</sub> uptake reported in this study has been previously reported for other ecosystems (Hartmann et al., 2011;Stiehl-Braun et al., 2011;Castro et al., 1994;Price et al., 2003). This agrees with the theory that soil CH<sub>4</sub> uptake is mainly limited by diffusion in most forest ecosystems (Price et al., 2004) when the sites of microbial CH<sub>4</sub> oxidation are distributed through the surface soil (Stiehl-Braun et al., 2011), and the concentration gradient between soil and atmosphere, which drives the flux, is effectively constant (von Fischer and Hedin, 2007). However, previous field studies have never been able demonstrate so conclusively the strength of the relationship (>90% variation explained) between AFP and soil CH<sub>4</sub> uptake, and for two separate forest systems. To our knowledge the only other study where similarly strong correlations between soil moisture and CH<sub>4</sub> uptake have been reported, was for grassland soils under summer rainfall exclusion (Hartmann et al. (2011).

It is important to note that WFPS has commonly been used to model, or compare, soil  $CH_4$  uptake in different ecosystems (Del Grosso et al., 2000). However, in our study this soil environmental variable was not as effective as AFP in explaining the observed  $CH_4$  flux patterns at the two temperate forest sites. At an individual site level, the relationship between WFPS and  $CH_4$  uptake had the same coefficient of determination as between AFP and  $CH_4$  uptake, however, the slope of the relationship differed between the two forest sites (Fig. 4D). This suggests that WFPS is not the most suitable soil moisture metric to relate soil gas diffusivity to soil  $CH_4$  flux when comparing sites or ecosystems. This is most likely due to the fact that WFPS is a proportional measure that relates VWC to the total soil porosity (equation (4)); compared to AFP that is a direct expression of the air filled pore volume in a given soil (equation (5)).

This demonstrates that soil gas diffusivity is primary related to the volumetric fractions of air (AFP), rather than the volumetric fraction of water in the soil since diffusion through air is much faster than through water (Farquharson and Baldock (2008)).

Our data also show a very weak influence by soil temperature upon soil CH<sub>4</sub> uptake. This temperature effect appears to be mainly driven by the correlation between soil moisture and soil temperature, which is typical for the climate of the investigated forest systems. After the effect of soil moisture was accounted for soil temperature was only able to account for less than 5% of the remaining variability in soil CH<sub>4</sub> flux at AU-WOM and less than 1.5% of the remaining variability in soil CH<sub>4</sub> flux at AU-WRR. Furthermore, the daily temperature variation in soil CH<sub>4</sub> uptake would have been masked in the analyses because all regression analyses were performed on either chamber cycle or daily uptake means. However, the overall weak but statistically significant temperature dependency of soil CH<sub>4</sub> uptake is unlikely to greatly influence seasonal CH<sub>4</sub> flux variability given that at both sites around 90% of seasonal variability in CH<sub>4</sub> uptake can be explained by soil moisture alone and that soil moisture and temperature are weakly correlated in the investigated forest systems. This was more pronounced at the AU-WOM site because temporal soil moisture variability was greater and we had two years of data compared to one year of data at the AU-WRR site. However, a model that includes soil temperature and soil moisture together performed marginally better based on the AIC as compared to a model that only used soil moisture status in predicting soil CH4 flux at both of our sites, which is logical based on the fact that all soil microbial processes show a physiological temperature response but it appears that for the MOB temperature response is rather muted at our sites during our measurement timeframe. Furthermore, our data also show that soil CH<sub>4</sub> uptake still continued at a very low WFPS of 10% (VWC = 0.07 g cm<sup>-3</sup>, AFP = 0.59 cm<sup>3</sup> cm<sup>-3</sup>) with CH<sub>4</sub> uptake ranging between -62 to -80  $\mu$ g CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> at this time. We can therefore hypothesize that MOB activity was not severely limited by moisture at the AU-WOM and the AU-WRR sites during the measurement period.

This study reports continuous measurement of soil-atmosphere CH<sub>4</sub> exchange in two temperate eucalypt forests in Australia measured at high temporal resolution for >12 month. Mean daily CH<sub>4</sub> flux values (AU-WRR = -1.35 to -0.12 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>; AU-WOM = -1.36 to -0.11 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) were well within the reported range for other temperate forests in Europe (-2.47 to + 0.26 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>; (Smith et al., 2000)) or worldwide (-10.68 to 0.02 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>; (Dalal et al., 2008;Dalal and Allen, 2008)).

The estimated annual CH<sub>4</sub> uptake of -1.79 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> for AU-WOM and -3.83 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> for AU-WRR are comparable to the range of -2.5 to - 3.7 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> reported for temperate beech and spruce forest sites in Germany where CH<sub>4</sub> fluxes were measured with a similar automated system over multiple years (Butterbach-Bahl and Papen, 2002). Globally, a range of -1.31 to -10.5 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> has been reported for temperate forest systems based on short and long-term, automated and manual chamber measurement campaigns (Dalal et al., 2008;Dalal and Allen, 2008). The annual CH<sub>4</sub> uptake rate estimated for AU-WOM in our study was less than a third of the -5.8 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> estimated by Meyer et al. (1997) for soils in the same forest system. This earlier CH<sub>4</sub> sink estimate was based on only five seasonal flux measurements but might also be attributed to the measurements being taken during three dry years (1993 – 1995) when average rainfall was 677 mm yr<sup>-1</sup> (Meyer et al. (1997). In comparison, the years when our study was undertaken (2010 – 2012) the average rainfall was 1063 mm yr<sup>-1</sup>. This may partly explain the greater CH<sub>4</sub> uptake estimate of Meyer et al. (1997) as the lower soil moisture levels may well lead to greater CH<sub>4</sub> uptake rates."

3) I found so many typographical errors throughout the manuscript.

We apologise for the large number of typographical errors Reviewer 1 discovered and we have endeavoured to correct all to greatly improve the presentation, writing and communication.

### **Abstract**

P1L13-14. Add "under predicted climate change scenarios" to the sentence.

## This has been added

P1L26. Replace "air-filled porosity" with "AFP" as the abbreviation appears in P1L21.

# This has been replaced

P1L23-25. I disagree with this statement after reading the results and discussion. Activity of MOB was not quantified in this study, and the results cannot indicate MOB activities were similar between the two sites.

We agree and have rephrased this sentence to now read as:

"Our data suggest that soil MOB activity in the two forests was similar and that differences in soil CH<sub>4</sub> exchange between the two forests were related to differences in soil moisture and thereby soil gas diffusivity."

P1L24. Check "physiochemical" for definition. It can be a typo for "physicochemical".

This word has been removed as a consequence of the rephrasing of the sentence above.

P1L24. Here, the differences between the two sites in CH4 flux were due to "physiochemical" but AFP explained up to 90% of the variability, indicating that the differences were likely caused by moisture regime.

Soil moisture is a physicochemical difference; it is not a biological difference. However, we have agreed to change the sentence and remove the reference to physicochemical and the sentence now reads as:

"Our data suggest that soil MOB activity in the two forests was similar and that differences in soil  $CH_4$  exchange between the two forests were related to differences in soil moisture and thereby soil gas diffusivity."

Introduction

P2L17. Here "air filled" is not hyphenated. Be consistent throughout the manuscript.

We have standardised this word to appear as "air-filled" throughout.

P2L33-34. This statement needs citation.

This statement has citations.

P2L35-P3L1. There are many ecosystems in the Northern Hemisphere without snow or below zero soil temperature, comparable to the Australian forests (e.g., Southwest of USA, Mediterranean region).

Reviewer 1 is correct in that there are bioregions in North America that have similar temperature ranges as Australian temperate forest soils. However, this statement is a follow up 'Furthermore,' statement to the primary one, which states that Australian temperate forest soils are highly weathered and very low in nutrients. No change has been made.

Results

P7L23. Replace "around" with "approximately"

Agreed, we have replaced.

P7L23. Fig. 4 should not appear before Fig. 3 (P8L1) in the text.

Agreed, we have changed the order of the text.

P7L23. The text mentions 85% (0.85), but Table 1 has 0.896. Why are they different? It's also the case for 19% in the text, and 0.148 in the table.

Table 1 has 0.855 for VWC and we rounded this to 85%. However the 19% was misquoted and we have corrected this to 16% as represented by and R2 of 0.158 in table 1 for soil temperature.

P7. This is not the best way to analyze the data. Use a model selection approach such as Akaike Information Criterion. For instance, see Monteith et al. (2015): Monteith DT, Henrys PA, Evans CD, Malcolm I, Shilland EM, Pereira M (2015) Spatial controls on dissolved organic carbon in upland waters inferred from a simple statistical model. Biogeochemistry 123(3): 363-377

We agree and have re-analysed the data as outlined above. Based on the AIC for each site a model using soil moisture and soil temperature performs marginally better compared to a model using soil moisture alone to predict soil  $CH_4$  uptake (see above). However, the additional amount of  $CH_4$  variance explained by including soil temperature into the model as compared to a model only including soil moisture is less than 1% at both sites. Including temperature in addition to moisture may on a statistically basis improve the model accuracy but not the predictive capacity. Hence, our overall conclusion is still valid. However, we agree that the AIC is the better way of selecting the best model.

P7L30. Avoid starting a sentence using an abbreviation. Spell out AFP.

Agreed - we have changed this throughout the manuscript.

P8L1. There are AU-Wom and AU-WOM, and AU-Wrr and AU-WRR. Stick to one form.

Agreed, we have now standardised these acronyms to all appear in CAPITALS

P8L20-21. This should be SD, not SE. The large sample size (how many?) makes the SE so small and misleading.

We have changed these to Standard Deviations rather than Standard Errors.

P8L7-9. Awkward sentence, and I cannot find "inter annual" differences were displayed well in figures.

Agreed, this sentence has been deleted. It effectively replicated, in a confusing way, the statement in the previous sentence and was therefore redundant.

P8L9. VWC in the text, but soil moisture in the figure. Be consistent.

We have standardised this to volumetric water content (VWC) throughout the manuscript where appropriate. We use soil moisture as a general term that stands for the different ways soil moisture can be expressed (AFP, VWC and WFPS).

P8L10. Fig. 4a in the text, and Fig. 4A in the figure. Be consistent.

We have standardised reference to figures with upper-case e.g. "Fig. 4A"

P8L28-P9L7. What is the point of presenting daily CH4 flux in relation to soil environmental variables, if it is not better than that in finer time scales, and does not add much?

We disagree with the reviewer since a lot of available flux studies only ever cite daily flux values and the relationships of daily flux values with environmental variables. We believe that the

inclusions of this information in the manuscript will especially be of interest to modellers and will help to put our data in the context of  $CH_4$  flux studies globally.

P9L5-8. Integrate this section to the first paragraph of the Results.

Given that the annual site  $CH_4$  flux budgets are calculated based on the daily flux data presented in the two preceding paragraphs of the result section, we believe it is more logical to leave this section where it is.

#### Discussion

P9L10. 1-2 years? I thought the measurements were for two years. Spell out numbers smaller than nine

We agree this is confusing. We have changed this to read ">12 months"

Start the discussion on the most exciting findings. I believe the significant correlation between AFP and CH4 flux for the two sites is most interesting in this study. Comparing the daily CH4 flux values with past studies is not too exciting.

Agreed. We have rearranged the Discussion sections to open with the discussion of AFP and CH<sub>4</sub> flux.

P9L25-31. Delete the paragraph. I do not think the statement is true that cool wet temperate eucalypt forests are often compared to rainforests. It's interesting that the annual CH4 flux is comparable between the eucalypt forest and a tropical rainforest, but no more than that. Plus, net CH4 flux is determined by not only MOB activities, but also methanogens as well, especially in wet sites. Thus, there is not much point for the comparison.

We agree with the reviewer and have removed this section form the discussion

P10L1-4. This paragraph should be presented first in the discussion.

We agree and now open the Discussion with this paragraph

P10L10-11. Check the order of the citations.

We have checked that our citation style is consistent with the BGS style format throughout the manuscript

P10L13-15. Is this an assumption? Delete (i.e. atmospheric CH4 concentration) and add "between soil and atmosphere" after "the concentration gradient" (L14).

# We have made this suggested change

P10L21-L23. The coefficient of determination for the relationship between WFPS and CH4 uptake is mentioned in the text, but not shown in Table 1 or 2. The relationship is shown in Fig. 4D, but the coefficient is not shown. If it's discussed in the text, it should be shown somewhere.

Agreed. We have added coefficients of determination to Figure 4

P10L28-29. Delete the sentence, and cite Farquharson and Baldock (2008) for the previous sentence.

Agreed, we deleted the last sentence of the paragraph and placed the citation after the previous sentence.

P10L35-P11L2. This sentence needs to be integrated into the context, otherwise it does not make sense. The paragraph is discussion about temperature on CH4 flux. Then, out of the blue, the sentence on CV of CH4 flux appears without relating it to temperature. It's confusing. I am not quite convinced that temperature did not affect CH4 flux with the current analyses. The better way to test the temperature effect is that 1) construct two models (CH4 flux is a function of moisture, and moisture + temperature) and 2) compare the two models via AIC. This will provide a more concrete answer.

In response to the reviewer's suggestion we have removed the sentence about the CV from the paragraph. In response to the second part of the reviewers comment please see our responses above. We have performed the suggested analysis with the result that the AIC indicates that at both sites models including soil temperature and soil moisture perform marginally better as compared to models only including soil moisture to explain CH<sub>4</sub> flux variability.

P11L2. Replace "will" with "would".

# Agreed, this change has been made

P11L9-14. I do not think the statement is valid. First, the authors measured soil moisture only to 10 cm in depth, and did not measure soil moisture in deeper soils. Methanotrophs in deeper soils can contribute to CH4 oxidation if the surface soils are dry. The only way to tease out methanotroph activity from physical constraints of soils for CH4 diffusion is to measure CH4 flux as well as gas diffusivity (see von Fischer et al. 2009). von Fischer JC, Butters G, Duchateau PC, Thelwell RJ, Siller R (2009) In situ measures of methanotroph activity in upland soils: A reactionâ AR diffusion model and field observation of water stress. Journal of Geophysical Research: Biogeosciences (2005–2012) 114(G1):

We have removed the strong statement that our data clearly demonstrate that there was no moisture limitation of MOB activity at the beginning of this paragraph and now simply make a statement that we didn't see any indication that soil  $CH_4$  uptake was moisture limited in our data. This paragraph it now reads:

"Furthermore, our data also show that soil  $CH_4$  uptake still continued at a very low WFPS of 10% (VWC = 0.07 g cm<sup>-3</sup>, AFP = 0.59 cm<sup>3</sup> cm<sup>-3</sup>) with  $CH_4$  uptake ranging between -62 to -80  $\mu$ g  $CH_4$  m<sup>-2</sup> h<sup>-1</sup> at this time. We can therefore hypothesize that MOB activity was not severely limited by moisture at the AU-WOM and the AU-WRR sites during the measurement period."

However, we also want to point out that the suggested test following the method described by von Fischer 2009 can not necessarily provide the information needed to assess if there is in-situ moisture limitation of methanotrophic activity because this method treats the soil as a one layer and provides a bulk methanotorphic activity measurement and a bulk soil diffusivity measurement without any information on where along the soil profile methanotrophic activity happens at any given point in time. Which means that if as a result of increased diffusivity a larger area of the profile compensates for lower MOB activity in the top soil layer (caused by moisture limitation) it will not show up.

P11L17. Why is "air filled porosity" used here, instead of AFP? Be consistent throughout the manuscript.

# Agreed, we have changed to AFP

P11L17. Replace "same" with "almost identical" (they are not "same" based on Table 1).

# Agreed, this change has been made

P11L18-19. I disagree with the statement. It is possible that AFP governs the CH4 flux across the landscape for eukalypt forests, but there is also a possibility that the casual correlations between AFP and CH4 flux happened to be very similar for the just two study sites. It's not reasonable to extrapolate the results to all the same type of forests in Australia.

Agreed - this statement was an over-reaching. We have reworded this to now read as:

"This means that future research should investigate whether simple information about soil bulk density can be used to estimate CH<sub>4</sub> uptake across different eucalypt forest ecosystems in Australia, or in other similar ecosystems globally."

**Tables** 

P16L5. "S" in "TS" should be subscript.

Agreed, we have made the subscript change in the Table caption.

Is "-" missing for 195.768 for the AFP parameter at AU-Wom?

#### Yes this has been corrected

"Soil water content" is used in the caption. Is this the same as "soil moisture content" (e.g. P30L6)? If so, use only soil water content.

We are using soil moisture content in the manuscript as a general term that represents the three different ways soil moisture content can be expressed (AFP, VWC and WFPS). We believe this is clear and not incorrect.

P17. Table 1. Are "constants" intercepts? Are "parameters" slopes for predictor variables?

Yes, to clarify this we have added a sentence to the caption.

Are both "unstandardized and standardized coefficients" in parentheses?

We have clarified this in the caption only standardized coefficients are in parentheses

Table 1 shows results of four regressions: 1. VWC 2. Soil temp 3. VWC and soil temp 4. AFP

But the corresponding Fig. 4. has: 1. VWC 2. AFP 3. Soil temp 4. WFPS Why the inconsistency?

The reasoning behind displaying WFPS in to  $CH_4$  flux relationship in Figure 4 was that we wanted to show that if WFPS is used as a measure of soil moisture the slopes of the relationships with  $CH_4$  flux are different at each site. We will add WFPS to the tables 1 and 2

**Figures** 

18. I am not sure if the data are presented in the most effective manner in Fig. 1 and 2. The current figures have; A: CH4 flux B: Air temp, CV of CH4 flux, and precip C: Soil temp and soil moisture Is there rationale behind the combinations? I am not convinced that the arrangement makes sense. How about rearrange the combinations; A: CH4 flux and CV B: Precip and soil moisture C. Air and soil temp

Or A: Ch4 flux B: Precip, soil moisture and CV C. Air and soil temp

We arranged the figures this way because the CV% of the  $CH_4$  fluxes would convolute figure A to a point where  $CH_4$  flux and CV% cannot be separated visually. We decided to leave the figure presentation as it stands.

P18. Fig. 1. AU-Wrr and AU-Wom are Fig. 1A and 1B, respectively, but "A" and "B" letters in the parentheses do not match up. Are these typo?

Yes, we have corrected this typographical error in the Figure caption

Replace SE with standard deviations. SE partly depends on the sample size, which is not described in the text, thus the tight error bars can be misleading.

Agreed, we have changed to SE to SD in Fig 1A and 1B

P19. Fig. 2. The description is confusing. Are the individual symbols means of measurements over four hours for each chamber, or average of 10 chambers? Spell out "four" instead of "4".

Agreed – this can be interpreted several ways. We have rewritten this line in the Figure caption to now read as:

"Soil-based flux of  $CH_4$  at a mixed Eucalyptus obliqua and E. regnans forest stand. Warra LTER, Tasmania, Australia (AU-WRR). Panel A shows  $CH_4$  flux cycle means of ten chambers measured within a four hour time period, panel B shows in black closed symbols site air temperature averaged over the chamber cycle period, daily rainfall sums (bars) and coefficient of variance of the  $CH_4$  flux cycle mean shown in Panel A (grey closed symbols). Panel C shows soil temperature in the top 0-10 cm averaged over each chamber cycle (grey open symbols) and corresponding volumetric soil water content (grey closed symbols) at the site

Replace "moisture" with "water".

In the text (P7 L20), it seems like CV was calculated using average and SD of 10 chambers, but it seems like CV was calculated using average and SD over time for each chamber.

We agree that this was unclear. CVs are calculated using average and SD of 10 chambers in one measurement period. We have changed the caption to:

"Soil-based flux of  $CH_4$  at a mixed Eucalyptus obliqua and E. regnans forest stand. Warra LTER, Tasmania, Australia (AU-WRR). Panel A shows  $CH_4$  flux cycle means of ten chambers measured within a four hour time period, panel B shows in black closed symbols site air temperature averaged over the chamber cycle period, daily rainfall sums (bars) and coefficient of variance of the  $CH_4$  flux cycle mean shown in Panel A (grey closed symbols). Panel C shows soil temperature in the top 0-10 cm averaged over each chamber cycle (grey open symbols) and corresponding volumetric soil water content (grey closed symbols) at the site.

P21. Fig. 4. Add regression equations and R<sup>2</sup> values on the figures."

# Agreed, the Figure has been annotated with regression equations and R2.

P22. Fig. 5. I do not think this is the best way to present the data. In the text, the authors want to show there is no significant correlation between CH4 flux and inorganic N contents. Then, scatter plots should be used to show the data.

We disagree, our intention was to give the reader an idea of the temporal variability in soil nitrate and ammonium concentrations, what can clearly only be achieved with a figure that can accommodate a timeline. The  $R^2$  and P values of the regressions between nitrate and ammonium concentrations and  $CH_4$  flux are listed in the caption of the figure. We believe that it is quite clear from the figures that there is no significant relationship between these parameters.

# **Reviewer 2**

The authors investigated the soil methane exchange at two Australian forest study sites differing in annual precipitation. Their major finding is that soil moisture is the main controlling factor and that the relationships of the two sites collapse if air-filled soil porosity, instead of water-filled pore space or volumetric soil moisture is used. The paper is of interest to the readers of BG and the main finding is of general interest to the community as it may trigger new approaches of simulating soil methane exchange if verified across a larger number of sites.

# I have three major comments:

(1) The study uses two different measurement systems at the two study sites. How can the authors ascertain that the two measurement systems do not cause systematic differences between the two sites? Without a cross-comparison between the two systems at the same site, how can we believe the differences/ lack of differences between sites when normalised with AFP?

We agree with the reviewer that this is an important point to consider. In our previous research over the last 15 years we have used different measurement systems in many ecosystems in Australia and some of them in the same ecosystem. When using closed-static and closed-dynamic systems in the same ecosystem we never detected large differences in the CH<sub>4</sub> flux magnitude. However, we have not been able to test the two automated systems in parallel at the same site. In fact such a comparison has not been conducted anywhere in the literature as far as we are aware. There have been many studies on chamber design and comparisons between automated and manual systems, mainly for  $CO_2$  and some for  $N_2O$ . But a systematic evaluation of an automated closed-static and an automated closed dynamic system for CH<sub>4</sub> flux has not been performed. Furthermore there are examples in the literature where data from different manual chamber systems (dynamic and static) and different automated chamber systems (static, dynamic, different analysers) where used a) to compare CH<sub>4</sub> flux magnitudes and b) to derive general functional relationships between soil temperature and moisture and soil CH<sub>4</sub>, fluxes across multiple ecosystems biomes and continents (Curry, 2009, 2007; Dalal and Allen, 2008; Dalal et al., 2008; Del Grosso et al., 2000; Smith et al., 2000). We do not see how our approach is any different. Hence, we acknowledge that it is possible that the two measurement systems could have measured different magnitude of  $CH_4$  flux in the two ecosystems and that by chance the relationship between CH<sub>4</sub> flux and AFP is identical at the two sites. However, we observed a very strong linear relationship between CH<sub>4</sub> flux and AFP at each site and AFP was able to predict around 90% of the flux variation. This is true regardless of the slope of the relationship. So the only difference that the measurement magnitude could make is a different offset of the slope of the relationship. Hence, we include following qualifying statement in the Discussion that highlights this possibility.:

"It is possible that the two different measurement systems (GC at AU-WRR and FTIR at AU-WOM) could produce different measures of  $CH_4$  flux if operated at the same site because of technological and methodological differences. If that were true, there would only be a remote chance that the two linear relationships between  $CH_4$  flux and AFP would overlap one another. As such, our finding that the relationships between  $CH_4$  flux and AFP do converge into one common regression line (as shown in Fig. 4) is worthy of note and suggests similar accuracy between the two measurement systems and similar function in soil  $CH_4$  exchange processes at the two forest sites."

Curry, C. L.: The consumption of atmospheric methane by soil in a simulated future climate, Biogeosciences, 6, 2355-2367, 2009.

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Dalal, R. C. and Allen, D. E.: Greenhouse gas fluxes from natural ecosystems, Australian Journal of Botany, 56, 369-407, 2008.

Dalal, R. C., Allen, D. E., Livesley, S. J., and Richards, G.: Magnitude and biophysical regulators of methane emission and consumption in the Australian agricultural, forest, and submerged landscapes: a review, Plant and Soil, 309, 43-76, 2008.

Del Grosso, S. J., Parton, W. J., Mosier, A. R., Ojima, D. S., Potter, C. S., Borken, W., Brumme, R., Butterbach-Bahl, K., Crill, P. M., Dobbie, K., and Smith, K. A.: General  $CH_4$  oxidation model and comparisons of  $CH_4$  oxidation in natural and managed systems, Global Biogeochemical Cycles, 14, 999-1019, 2000.

Smith, K. A., Dobbie, K. E., Ball, B. C., Bakken, L. R., Sitaula, B. K., Hansen, S., Brumme, R., Borken, W., Christensen, S., Prieme, A., Fowler, D., Macdonald, J. A., Skiba, U., Klemedtsson, L., Kasimir-Klemedtsson, A., Degorska, A., and Orlanski, P.: Oxidation of atmospheric methane in Northern European soils, comparison with other ecosystems, and uncertainties in the global terrestrial sink, Global Change Biology, 6, 791-803, 2000.

(2) The results of the two sites should be presented together instead of separately for each site.

We disagree with this suggestion as it is important not to suggest to the readers that these sites can be directly compared. They are independent sites that simply show similar strength of relationship between AFP and CH<sub>4</sub> flux. Furthermore, they span difference time periods, experience difference rainfall conditions which would make the figures far more confusing and with less visual resolution.

(3) English style and grammar are in the need of checking by a native speaker.

Agreed. We have extensively edited the manuscript to improve the presentation communication and grammar. Comments from Reviewer 1 greatly assisted in this.

Detailed comments:

p. 1, 1. 13-14: this sentence comes a bit as a surprise

We have added following sentence ahead of this sentence:

"Soils in temperate forest ecosystems are the greatest terrestrial  $CH_4$  sink globally."

p. 2, 1. 2: high compared to many VOC that are present in the ppt range ...

Reviewer 2 is correct. We have changed this sentence to now read as:

"Methane (CH<sub>4</sub>) has an atmospheric concentration of  $\sim$ 1.8 ppm as compared to >400 ppm for carbon dioxide (CO<sub>2</sub>) the second most......"

p. 2, 1. 24: Q10 values critically depend on the depth of the soil temperature used as a reference due to increasing dampening in amplitude and phase shift with soil depth

Reviewer 2 is correct. However, the statement still holds as we only build upon and refer to the published literature.

p. 2, 1. 28: initiate a new paragraph here

Agreed, we have added a paragraph start.

p. 2, 1. 15-17: would the authors be able to formulate some hypothesis regarding their research? this would strengthen the paper

Our study was objective driven and we do not believe it would be correct to retrospectively add hypothesis to the introduction that would be based on the outcome of the study itself. The dataset is very strong and we firmly believe that the paper as presented it a very valuable contribution this field of research.

p. 3, 1. 28: density

Yes - we corrected this

p. 7, 1. 8: did you check for linearity of tested relationships?

Yes, linearity was critical acceptance of chamber flux data. We used an  $\mathbb{R}^2$  threshold of 0.9 for our quality control.

p. 7, l. 17: how many longer gaps did you encounter at both sites?

The figures clearly show when we encountered long data gaps at each site. However, we added a paragraph to the method section outlining the instrument failure related data gap percentage.

The paragraph reads:

"As outlined above, we excluded fluxes where the coefficient of determination of the regression of chamber concentration versus time was less than 0.9, which lead to the exclusion of approximately 10% of measured chamber fluxes. However, longer gaps in flux data, as shown in Figures 1A and 2A, are either a result of power failures or the need to switch of the power generators on days of extreme fire danger. This led to data gaps of around 30% of the individual datasets."

p. 7, 1. 29-30: figures should be reference in chronological order, i.e. Figure 3 after Figure 2

Agreed, we have changed the order of the Figures.

Results section: the paper would be much more easily readable, if the results of the two sites would be presented together, instead separately – this would help making a stronger point of the major finding of this study; this argument also applies to Figures 2 and 3, which should be combined in my view

We disagree that combining the results of the two sites would greatly assist the reader in understanding the data. We believe that it is important for the reader to clearly see that the relationship between environmental variables and  $CH_4$  flux is identical at each site – not to compare between the two sites. We have trialled this in a previous draft and it was less convincing.

p. 10, 1. 20: diffusion-limited

We have rephrased this to read as: "limited by diffusion..."

p. 10, l. 22: able to demonstrate p.

We rephrased this sentence to:

"This agrees with the theory that soil  $CH_4$  uptake is mainly limited by diffusion in most forest ecosystems (Price et al., 2004) when the sites of microbial  $CH_4$  oxidation are distributed through the surface soil (Stiehl-Braun et al., 2011), and the concentration gradient between soil and atmosphere, which drives the flux, is effectively constant (von Fischer and Hedin, 2007)."

10, 1. 31-33: reformulate in proper English

This sentence now reads:

This is most likely due to the fact that WFPS is a proportional measure that relates VWC to the total soil porosity (equation (4)); compared to AFP that is a direct expression of the air filled pore volume in a given soil (equation (5)).

p. 11, l. 3: I do not get the "However"

Agreed. We have deleted "However" at the start of this sentence.

p. 11, l. 11: what does "defined role" mean?

We have reworded this sentence to now read as:

"However, the weak temperature dependency of soil  $CH_4$  uptake is unlikely to greatly influence seasonal variability given that a..."

# Soil methane oxidation in both dry and wet temperate eucalypt forests show near identical relationship with soil air-filled porosity

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Abstract. Well-drained, aerated soils are important sinks for atmospheric methane (CH<sub>4</sub>) via the process of CH<sub>4</sub> oxidation by methane oxidising bacteria (MOB). This terrestrial CH<sub>4</sub> sink may contribute towards climate change mitigation, but the impact of changing soil moisture and temperature regimes on CH<sub>4</sub> uptake is not well understood in all ecosystems. Soils in temperate forest ecosystems are the greatest terrestrial CH<sub>4</sub> sink globally. Under predicted climate change scenarios, tTemperate eucalypt forests in south-eastern Australia are predicted to experience rapid and extreme changes in rainfall patterns, temperatures and wild fires. To investigate the influence of environmental drivers on seasonal and inter-annual variation of soil-atmosphere CH<sub>4</sub> exchange we measured soil-atmosphere CH<sub>4</sub> exchange at high temporal resolution (<2 hr) in a dry temperate eucalypt forest in Victoria (Wombat State Forest, 870 mm yr<sup>-1</sup>) and in a wet temperature eucalypt forest in Tasmania (Warra LTER, 1700 mm yr<sup>-1</sup>). Both forest soil systems were continuous CH<sub>4</sub> sinks of -1.79 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> in Victoria and -3.83 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> in Tasmania. Soil CH<sub>4</sub> uptake showed substantial temporal variation and was strongly controlled by soil moisture at both forest sites. Soil CH<sub>4</sub> uptake increased when soil moisture decreased, and this relationship explained up to 90% of the temporal variability. Furthermore, the relationship between soil moisture and soil CH<sub>4</sub> flux was near identical at both forest sites when soil moisture was expressed as soil air-filled porosity (AFP). Soil temperature only had a minor influence on soil CH<sub>4</sub> uptake. Soil nitrogen concentrations were generally low, and fluctuations in nitrogen availability did not influence soil CH<sub>4</sub> uptake at either forest site. Our data suggest that soil MOB activity in the two forests was similar and that differences in soil CH4 exchange between the two forests were related to differences in soil moisture and thereby soil gas diffusivity. Our data indicate that soil MOB activity in the two forests was similar and that differences in soil CH<sub>4</sub> exchange between the two forests were related to physiochemical differences in soil properties influencing soil gas diffusivity. The differences between forest sites and the variation in soil CH<sub>4</sub> exchange over time could be explained by soil air filled porosityAFP as an indicator of soil moisture status.

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#### 1 Introduction

Methane (CH<sub>4</sub>) has a relatively low atmospheric concentration of approximately 1.8 ppm and is after carbon dioxide (CO<sub>2</sub>, approx. 402 ppm) the second most abundant greenhouse gas in the atmosphere (IPCC, 2013). Although its atmospheric concentration is two orders of magnitude lower than that of CO<sub>2</sub>, CH<sub>4</sub> accounts for approximately 18% of the currently observed global temperature increase (IPCC, 2013). In addition, CH<sub>4</sub> contributes to 32% of the current radiative forcing created by the major greenhouse gases as it has a 25 times greater global warming potential (GWP) compared to CO<sub>2</sub> (IPCC, 2013).

Forest soils are the most important land based sink for CH<sub>4</sub> via the activity of methane oxidising bacteria (MOB) in well-drained, aerobic soils. Soils in temperate forest ecosystems play an important role in global CH<sub>4</sub> exchange between the land mass and the atmosphere, and they constitute around 30-50% of the soil based CH<sub>4</sub> sink worldwide (Ojima et al., 1993;Dutaur and Verchot, 2007).

Major environmental factors controlling and influencing CH<sub>4</sub> uptake rates by forest soils are soil diffusivity and -structure, soil moisture, soil temperature and soil nitrogen status (Ball et al., 1997a;Smith et al., 2003;von Fischer and Hedin, 2007;Butterbach-Bahl et al., 2002;Del Grosso et al., 2000).

The main factor regulating the CH<sub>4</sub> uptake capacity of soils is the diffusion rate of CH<sub>4</sub> through the soil and hence the substrate availability of CH<sub>4</sub> to the MOB across the soil profile. CH<sub>4</sub> uptake rates have been shown to decrease with increasing soil moisture as a result of decreasing soil gas diffusion rates across different ecosystems (Castro et al., 1995;Khalil and Baggs, 2005;Ball et al., 1997). Therefore, CH<sub>4</sub> uptake is thought to be most rapid in coarse-textured forest soils with a well-developed structure and an organic surface layer that does not inhibit gas diffusion (Boeckx et al., 1997;Del Grosso et al., 2000;Smith et al., 2000). Soil bulk density can also correlate with soil CH<sub>4</sub> uptake across different ecosystems (Smith et al., 2003;Smith et al., 2000), which is not unexpected since soil air-filled porosity, which is directly linked to soil diffusivity, is a function of soil bulk density and volumetric water content.

Soil CH<sub>4</sub> uptake at atmospheric levels generally shows limited temperature dependency and reported Q<sub>10</sub> values are generally low with an average around 1.4 (Crill, 1991;Born et al., 1990;Smith et al., 2000). Another factor that influences the CH<sub>4</sub> uptake capacity of soils is soil N status, especially the availability of ammonium (NH<sub>4</sub><sup>+</sup>) (Butterbach-Bahl et al., 1998;Sitaula et al., 1995). Increasing soil N availability through organic and inorganic fertiliser additions and through biological N fixation can decrease CH<sub>4</sub> uptake rates (Niklaus et al., 2006;Dick et al., 2006).

Temperate eucalypt (broadleaved evergreen) forests in south-eastern Australia cover around 26 million hectares (Committee, 2013), and provide a large range of ecosystem services. However, despite a growing interest in soil CH<sub>4</sub> uptake in the last decade there have been very few studies investigating CH<sub>4</sub> oxidation in soils of natural Australian forest and woodland ecosystems with only a relatively small number of published studies on CH<sub>4</sub> uptake in temperate forest systems (Fest et al., 2009;Livesley et al., 2009b;Meyer et al., 1997;Fest, 2013;Fest et al., 2015b;Fest et al., 2015a), tropical forest systems (Kiese et al., 2003) and savanna ecosystems (Livesley et al., 2011). Moreover, there is currently no model that accurately predicts

the size of the terrestrial CH<sub>4</sub> sink in Australia or determines how the strength of this sink will change over time. Data describing CH<sub>4</sub> emission and oxidation from Australian soils is still patchy and often lacking for important landscapes such as tropical savannas, the semi-arid and arid zones and woody ecosystems (Dalal et al., 2008).

Compared to most European and North American temperate forest systems, forest soils in the Australian temperate region are generally highly weathered and very low in nutrients, and atmospheric nitrogen deposition is very low. Furthermore most of the temperate forest area in Australia does not experience periods of snow cover or below zero soil temperatures. It is therefore questionable as to whether information gathered on spatial and temporal variability of soil CH<sub>4</sub> exchange in Northern Hemisphere temperate forest soils are transferable to those in Australia. Furthermore, it is not clear if processes that explain soil CH<sub>4</sub> uptake in deciduous forest systems or coniferous forest systems worldwide can be directly transferred to the eucalypt or acacia forest systems that dominate the forests and woodlands of Australia. Most estimates of soil CH<sub>4</sub> exchange in Australian forest systems were based on infrequent (weekly-monthly) or campaign-based measurements (of one to two weeks), which may not fully reflect the temporal dynamics and range of environmental conditions.

This study investigates soil-atmosphere CH<sub>4</sub> exchange using automated chamber systems measuring at a high temporal resolution over 1-2 years in two temperate *Eucalyptus obliqua* dominated forests sites with contrasting annual precipitation. The main objectives of this study were to assess the magnitude and temporal variation in CH<sub>4</sub> exchange between the soil and atmosphere in temperate evergreen eucalypt forest systems and to investigate the primary biophysical processes that control

#### 2 Material and Methods

the seasonality in soil CH<sub>4</sub> flux.

#### 2.1 Site description

The Tasmanian site is in the Warra Long Term Ecological Research (LTER) Site approximately 60 km west-southwest of Hobart, Tasmania, Australia (AU-Wrr: 43° 5'36.78''S, 146° 38'42.65''E). The site is dominated by *Eucalyptus obliqua* (L'Herit.) with an overstorey height of around 53 m and a basal area of 54 m² ha¹. The understorey is mainly comprised of *Acacia melanoxylon* (R.Br.), *Nothofagus cunninghamii* (Hook.) Oerst. and *Dicksonia antarctica* (Labill.). The climate of AU-Wrr-WRR is classified as temperate cool wet (Dunlop and Brown, 2008) with cold and wet winters and warm and wet summers. The average rainfall is approximately 1700 mm yr¹ (Fig. 1a1A) with mean monthly maximum temperatures of 19.3 °C in January (summer) and mean minimum temperatures of 2.5 °C in July (winter). The soils at Warra are derived from Permian siltstone with a surface texture of silty loam to silty clay loam, and are classifed as kurosolic redoxic hydrosol (McIntosh, 2012). The average bulk denisity in the top 5 cm of mineral soil is 0.67 g cm⁻³ and soil porosity is 0.74 cm³ cm⁻³. The Victorian forest site is in the Wombat State Forest, approximately 120 km west of Melbourne, Australia (AU-WomWOM: 37° 25'20.83''S, 144° 5'38.63''E). AU-Wom-WOM is dominated by *Eucalyptus obliqua* (L. Her.), *Eucalyptus rubida* (H. Deane & Maiden) and *Eucalyptus radiata* (Sieber ex DC) trees of approximately 20 – 25 m in height and 37 m² ha⁻¹ of stem basal area. The climate is classified as Mediterranean to cool temperate, with warm and dry summers and wet

and cool winters. The average rainfall is approximately 870 mm yr<sup>-1</sup> (Fig. 1b1B,) with mean monthly maximum temperatures of 25.6 °C in January (summer) and mean minimum temperatures of 3.4 °C in July (winter). The soils of AU-Wom-WOM are derived from weathered sandstone and shale, with a surface texture of sandy clay loam, classified as an acidic-mottled, dystrophic, yellow Dermosol (Robinson et al., 2003). The average bulk density in the top 5 cm of mineral soil is 0.90 g cm<sup>-3</sup> and soil porosity is 0.65 cm<sup>3</sup> cm<sup>-3</sup>.

# 2.1.1 Experimental design AU-WrrWRR

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The temporal variation in soil-atmosphere exchange of CH<sub>4</sub> was monitored continuously from 10/10/2010 to 15/01/2012 using a fully-automated gas chromatograph (GC) measurement system attached to ten pneumatic open-and-close chambers as described in Livesley et al. (2009). This system was supported by a remote area power system consisting of a 5kV diesel generator and 12V battery bank. The ten chambers were randomly distributed over an area of approximately 25 x 25 m. Chambers were attached to a square steel frame base (e.g. 50 cm x 50 cm) which was inserted 5 cm into the soil, and a plexiglass headspace of 15 cm depth (e.g. 37.5 L chamber volume). Chambers were attached to the frame using clamps and closed cell foam. For each chamber, six flux rate measurements were made during a 24 hour period, one every four hours. Further details of the automated trace gas measurement system, chamber design and gas chromatograph can be found in Butterbach-Bahl et al. (1997);Papen and Butterbach-Bahl (1999) and Livesley et al. (2009). Soil temperature (12-Bit Temp Smart Sensor, Onset Computer Cooperation, USA) was logged at 0-10 cm on a half hourly basis (Hobo U30, Hobo Data Logger, Onset Computer Cooperation, USA) in the middle of the site. Chamber pneumatic lids opened automatically when rainfall, measured by a tipping bucket rain gauge, exceeded 1 mm in 5 minutes to avoid a potential reduction in soil moisture inside the chambers caused by the rainfall exclusion during the relatively long time of chamber closure (2h).

# 2.1.2 Experimental design AU-WomWOM

Temporal variation in soil-atmosphere exchange of CH<sub>4</sub> was monitored continuously from 1/5/2010 to 30/04/2012 using a fully-automated Fourier Transform Infrared (FTIR) spectrometer measurement system attached to six pneumatic open-and-close chambers (Griffith et al 2012). This system was supported by a remote area power system consisting of a 4.5kV diesel generator and 24V battery bank. The automatic chambers used followed the same design as that described at the AU-Wrr WRR site. The opening and closing of the lids via pneumatic pistons was controlled with the measuring software on site (PC). Six chambers were distributed randomly over an area of around 25 x 25 m and were measured in sequence with each chamber initially having a measuring period of 15 minutes (1/5/2010 – 21/10/2010) that was later extended to 20 minutes to increase detection precision for other simultaneously measured trace gases (22/10/2010 – 30/04/2012). Lids were open for the first 2 and the last 2 minutes of every 15/20 minute measuring interval per chamber to flush the sample lines with ambient air resulting in a chamber incubation period of 11/16 minutes. One CH<sub>4</sub> flux measurement per chamber was achieved every 1.5/2 hours. The chambers were not fitted with a fan, but there was forced ventilation during the incubation

period of each chamber through the use of an external pump which circulated the air in a closed loop through the head-space of the chamber (closed dynamic setup), attached airlines (0.3 L tubing volume) and the measuring cell (3.5 L cell volume) of a Fourier Transform Infrared (FTIR) spectrometer setup (Spectronus, ECOTECH P\L, Australia). The spectrometer (Bruker IRcube with globar source and thermoelectrically cooled MCT detector) measured concentrations of CH<sub>4</sub>, CO<sub>2</sub>, N<sub>2</sub>O, carbon monoxide and water vapour in the air stream (Meyer et al., 2001;Griffith et al., 2012;Hammer et al., 2012). Measurements of the CH<sub>4</sub> concentration were made every minute during the 15/20 min chamber period. Further information about measuring principle, instrument setup, maintenance and calibration can be found in Griffith et al. (2012). Soil temperature (Thermocouple Probe) and moisture (impedance probes, ML2x – Theta Probe Soil Moisture Sensor, Delta-T Devices LTD, UK) was recorded continuously at 0-5 cm within each chamber. In addition, soil temperature (Averaging Soil Thermocouple Probe, TCAV, Campbell Scientific, Australia, Pty Ltd) and soil moisture (Water Content Reflectometer, CS616, Campbell Scientific, Australia, Pty Ltd) were recorded on a half hourly basis at 0-10 cm by an onsite eddy covariance system. Given the relatively short closure period of 11/16 minutes for each chamber during a 4 hour period, we decided that automated chamber opening in response to rainfall events was not necessary.

#### 2.2 Flux calculation

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15 CH<sub>4</sub> flux rates were calculated for both automated measuring systems from the rate of increase/decrease of gas concentration in the chamber head space with time according to:

$$F_{\mu L} = (V/A) x (dC_{CH4}/dt)$$
 (1)

Where V is the volume (L) of the chamber head space plus sample lines and the FTIR sample cell, A is the soil surface area covered by the chamber (m²) and t is time. The term  $dC_{CH4}/dt$  ( $\mu$ L L¹ h¹) was calculated from the initial linear CH<sub>4</sub> concentration change after chamber closure. In cases where the fitted linear regression model had an R² < 0.9 then this flux measurement was excluded from further analysis\_—The determined flux rate (F $_{\mu}$ L) was subsequently converted to  $\mu$ mol CH<sub>4</sub> m⁻² h⁻¹ (F $_{\mu}$ mol) by accounting for temperature, pressure and volume using Equation (2) based on the ideal gas law:

$$F_{\mu mol} = (F_{\mu L} x P) / (R x T)$$

$$(2)$$

Where *P* is the atmospheric pressure in kPa at site according to altitude or direct measurement (Eddy tower), *R* is 8.3144 (the ideal gas constant in L kPa<sup>-1</sup> K<sup>-1</sup>), and *T* is the air temperature in Kelvin (273.15 +  $^{\circ}$ C). Fluxes in  $\mu$ mol CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> were then converted to  $\mu$ g CH<sub>4</sub>-C m<sup>-2</sup> h<sup>-1</sup> based on the molecular atomic mass.

#### 2.3 Additional measurements

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From within each site, composite soil samples (three 0-5 cm samples) were collected, sieved (2 mm) and sub-sampled for 1M KCl extraction (1:4, soil:KCl) and gravimetric water content (GWC<sub>S</sub>) determination (105°C for 48 hours) during additional seasonal measurement campaigns spread across the measurement timeframe (n = 13 in AU-WomWOM, n = 10 in AU-WRRFF). KCl extracts were filtered (Whatman 42) and frozen prior to analysis for nitrate (NO<sub>3</sub><sup>-</sup>) and ammonium (NH<sub>4</sub><sup>+</sup>) concentration using an auto-analyser (SFA, Technicon TM).

During initial site installation (and over the course of the measurement timeframe) approximately 30 volumetric soil cores (0-5 cm, Ø 72 mm) were sampled at each site to determine soil volumetric water content (VWC) and soil bulk density (BD). The data were used to establish site dependent calibration curves between the onsite installed soil moisture sensors (HOBO Micro Station Data logger H21 and EC-5 Soil Moisture Smart Sensor, Onset Computer Corporation, USA), hand held impedance probes (ML2× Theta probe and HH2 Moisture Meter, Delta-T Devices Ltd, UK) and VWC (Kaleita et al., 2005). The bulk density and volumetric water content data and their relationship to the onsite installed soil moisture sensor readings and hand held impedance probes readings were further used to calculate soil porosity, air-filled porosity and percentage water filled pore space (%WFPS) for each plot and measuring event according to Loveday and Commonwealth Bureau of Soils (1973):

Soil porosity = 
$$1$$
- (soil bulk density / particle density) (3)

Where a value of 2.65 was used for particle density (g cm<sup>-3</sup>) of rock, sand grains and other soil mineral particles.

%WFPS = (volumetric water content 
$$\times$$
 100) / Soil porosity (5)

At the end of the study, a composite soil sample from five soil cores was collected at 0-5 cm at each site, air dried, sieved (2 mm) and analysed for soil particle size analysis through dispersion, suspension, settling and sequential hydrometer readings (Ashworth et al., 2001). A sub-sample of each air-dried soil was analysed for pH (1:5, soil:water) and for total C and N content using an elemental analyser (LECO®).

#### 2.4 Data presentation and statistical analyses

Flux and environmental sensor data presented (if not specifically related to individual chambers) in the figures here after are averages for respective chamber cycles where at least 2/3 of the chamber flux measurements had passed the above mentioned flux quality control (1.5/2 hour cycle for the FTIR system and a 4 hour cycle average for the GC system) at each

site ±1 SE (where error bars are present). We also calculated the coefficient of variance per chamber cycle (CV%<sub>cycle</sub>) by dividing the standard deviation of each chamber cycle by its respective mean and multiplying the result by 100. Furthermore, soil temperature and soil moisture data were averaged accordingly for each chamber cycle to allow regression analysis. In a second step, to enable correlation analysis with daily rainfall and sporadic soil inorganic nitrogen measurements we calculated daily site averages of the measured fluxes and environmental parameters, with the exception of rainfall where we calculated daily sums, for days where at least 80% of chamber cycles were available. We additionally calculated the coefficient of variation per day (CV%<sub>day</sub>) for the CH<sub>4</sub> flux data. As outlined above, we excluded fluxes where the coefficient of determination of the regression of chamber concentration versus time was less than 0.9, which lead to the exclusion of approximately 10% of measured chamber fluxes. However, longer gaps in flux data, as shown in Figures 1A and 2A, are either a result of power failures or the need to switch of the power generators on days of extreme fire danger. This led to data gaps of around 30% of the individual datasets.

All statistical analyses were performed with SPSS 20 (IBM, USA). Linear regression procedures and multiple linear regression procedures were used to investigate temporal relationships between measured soil environmental parameters and soil CH<sub>4</sub>. We initially ran stepwise linear regression procedure as an exploratory tool to identify significant predictors and predictor combinations and retested these afterwards in simple or multiple linear regression models. We transformed data when necessary to reduce heteroscedasticity for linear regression analysis. We used a restricted maximum likelihood framework (REML, automatic linear modelling in SPSS) to arrive at the Akaike information criterion for three selected models that predict soil CH<sub>4</sub> uptake (one model containing only soil temperature, one model containing only a measure of soil moisture (we choose AFP) and one model containing soil temperature and AFP as a predictors of soil CH<sub>4</sub> flux).

## 0 2.5 Annual site CH<sub>4</sub> flux budgets

To calculate annual site CH<sub>4</sub> flux budgets for both sites we first selected a 12 month period with the greatest data coverage for daily average flux for both sites (1/1/2011 – 1/1/2012) and filled existing flux data gaps as follows. For small data gaps of single days where no environmental sensor or flux data were available, we calculated values based on linear interpolation between the CH<sub>4</sub> flux of the day before the gap and the day after the gap. For data gaps longer than one day, we used the linear regression model between soil VWC soil moisture and daily soil CH<sub>4</sub> flux for each site (Table 1) to estimate the missing CH<sub>4</sub> flux data.

#### 3 Results

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#### 3.1 CH<sub>4</sub> flux in relation to soil environmental variables

At the AU-Wrr-WRR site, soil CH<sub>4</sub> flux was always negative indicating CH<sub>4</sub> uptake all year around (Fig. 2). The measurement cycle means ranged between -2  $\mu$ g CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> (spring 2010) to -58.4  $\mu$ g CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> (autumn 2011) with an arithmetic mean of -41.2  $\pm$  0.2311.0 SE-SD  $\mu$ g CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>. In general, months with higher average soil moisture and higher

total rainfall displayed lower CH<sub>4</sub> uptake when compared to months with lower average soil moisture and lower total rainfall (Fig. 2). Inter annual differences in average site CH<sub>4</sub> uptake between seasons (spring and summer) were also reflected in concurrently recorded average site soil moisture levels. The coefficient of variance (CV) for the average CH<sub>4</sub> flux based on 10 chambers in one measurement cycle ranged between 1.8 and 98.0% with an average of 17.9  $\pm$  0.2311% (SESD) and was higher in periods of rapid changes in soil moisture levels reflecting changes in precipitation (Fig. 2).

At the AU-WOM site soil  $CH_4$  flux was always negative, indicating  $CH_4$  uptake all year around (Fig. 3). The measurement cycle means ranged between -1.3  $\mu$ g  $CH_4$  m<sup>-2</sup> h<sup>-1</sup> (recorded during a period of heavy rainfall in summer 2011) to -62.5  $\mu$ g  $CH_4$  m<sup>-2</sup> h<sup>-1</sup> (summer 2010) with an arithmetic mean of -25.5  $\pm$  12.7 SD  $\mu$ g  $CH_4$  m<sup>-2</sup> h<sup>-1</sup>. Similar to the AU-WRR site, months with higher average soil moisture and higher total rainfall displayed lower  $CH_4$  uptake when compared to months with lower average soil moisture and lower total rainfall (Fig. 3). The CV for the average  $CH_4$  flux based on six chambers in one measurement cycle ranged between 6.7 and 143.0% with an average of 29.3  $\pm$  9.7% (SD) and was again higher in times of rapid soil moisture changes in response to changes in precipitation patterns (Fig. 3).

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For AU-WRR the linear regression analysis showed that volumetric water content (VWC) accounted for around approximately 85% of variability in soil CH<sub>4</sub> uptake across all seasons (Fig. 4a4A, Table 1) with soil CH<sub>4</sub> uptake decreasing when soil VWC increased or soil CH<sub>4</sub> uptake increasing when air-filled porosity (AFP) increased (Fig. 4b4B, Table 1). Soil temperature (0-5 cm) alone was weakly related to CH<sub>4</sub> uptake with higher CH<sub>4</sub> uptake rates associated with higher soil temperatures. However, soil temperature alone was only able to account for approximately around 1916% of seasonal variability in CH<sub>4</sub> uptake (Fig. 4e4C, Table 1). In addition, after taking the effect of VWC into account, soil temperature only explained around 1.5% of the remaining variability in CH<sub>4</sub> uptake at AU-Wrr-WRR (data not shown). A regression model containing VWC and soil temperature as input variables had only a marginally higher coefficient of determination when compared to the model only containing VWC (Table 1). AFP-Air-filled porosity or VWC showed some weak dependency of soil temperature at the site (R<sup>2</sup> = 0.14, p < 0.001).

At the AU WOM site soil CH<sub>4</sub> flux was also always negative indicating CH<sub>4</sub> uptake all year around (Fig. 3). The measurement cycle means ranged between 1.3 μg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> (recorded during a period of heavy rainfall in summer 2011) to -62.5 μg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> (summer 2010) with an arithmetic mean of -25.5 ± 0.16 SE μg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>. Similar to the AU WRR site months with higher average soil moisture and higher total rainfall displayed lower CH<sub>4</sub> uptake when compared to months with lower average soil moisture and lower total rainfall (Fig. 3). The CV for the average CH<sub>4</sub> flux based on 6 chambers in one measurement cycle ranged between 6.7 and 143.0% with an average of 29.3 ± 0.12% (SE) and was again higher in times of rapid soil moisture changes in response to changes in precipitation patterns (Fig. 3). Furthermore, interannual differences in average site CH<sub>4</sub> uptake between seasons were also reflected in concurrently recorded average site soil moisture levels. For AU-WOM the linear regression analysis showed that VWC could account for around 91% of variability in soil CH<sub>4</sub> uptake across all seasons (Fig. 4a4A, Table 1) with soil CH<sub>4</sub> uptake decreasing when soil VWC increased, the opposite trend was observed for AFP (Fig. 4b4B, Table 1). Soil temperature (0-5 cm) alone was again weakly related to CH<sub>4</sub>

uptake with higher CH<sub>4</sub> uptake rates associated with higher soil temperatures (Fig. 4e4C). At the AU-WOM site, only around 20% of seasonal variability in CH<sub>4</sub> uptake (Table 1) was explained by soil temperature. In addition, similar to the results at AU-WRRFF, after taking the effect of VWC into account, soil temperature only explained around 5% of the remaining variability in CH<sub>4</sub> uptake at AU-Wom-WOM (data not shown). Furthermore, a regression model containing VWC and soil temperature had a marginally lower coefficient of determination (Table 1) when compared to the model only containing VWC (Table 1). AFP-Air-filled porosity or VWC showed some weak dependency of soil temperature at the site (R<sup>2</sup> = 0.38, p < 0.001).

The AIC results of the REML analysis confirm the results of the linear regression approach (Table 2) showing that soil moisture (in this case expressed as AFP) is the strongest predictor of soil CH<sub>4</sub> flux in both forest systems. The analysis shows that the models including soil moisture and soil temperature perform marginally better based on AIC compared to models including only soil moisture to predict soil CH<sub>4</sub> flux. However, the importance rating of the predictors (soil moisture and soil temperature) clearly indicates that in both forest systems soil moisture dominates, as it accounts for more than 99% of the proportion of variance explained by the model compared to <0.01% proportion of the variance explained by soil temperature.

# 3.2 Mean daily and annual CH<sub>4</sub> flux in relation to environmental variables

## 3.2.1 Site AU-WrrWRR

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Daily site averages ranged between -0.12 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> and -1.35 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> with an arithmetic mean of -0.98  $\pm$  0.2702 SDE mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. The coefficient of determination for the regression analysis changed slightly when the regression analysis were calculated on daily means and VWC was able to account for up to 89% in the observed variability in CH<sub>4</sub> flux (Table 23). The CV for the daily average site CH<sub>4</sub> flux ranged between 0.15% and 20.6% with an average of 3.5  $\pm$  03.33.19% (SDE) and was higher in periods of rapid changes in soil moisture levels. We calculated soil CH<sub>4</sub> flux averages for 3 days around the dates when soil NH<sub>4</sub><sup>+</sup> and soil NO<sub>3</sub><sup>-</sup> samples were taken on-site to enable regression analysis; however, neither NH<sub>4</sub><sup>+</sup> nor NO<sub>3</sub><sup>-</sup> alone or together could explain any variability in soil CH<sub>4</sub> flux at the site and all relationships were non-significant (Fig. 5b5B, dD, fF).

# 3.2.2 Site AU-WomWOM

Daily site averages ranged between -0.11 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> and -1.36 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> with an arithmetic mean of -0.62  $\pm$  0.010.30 SdE mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. The CV for the daily average site CH<sub>4</sub> flux ranged between 0.11% and 47.6% with an average of 5.6  $\pm$  4.360.17% (SESD) and was again higher in periods of rapid changes in soil moisture levels. As for the AU-WfF WRR site the coefficient of determination for the regression analysis changed slightly when the regression analysis was calculated on daily means and VWC was able to account for up to 92% in the observed variability in CH<sub>4</sub> flux (Table 23).

Similar to the AU-Wrr-WRR site, three day CH<sub>4</sub> flux averages were not significantly correlated with soil  $NH_4^+$  or  $NO_3^-$  if entered alone or together as predictors to the linear regression model (Fig. 5a5A, eC, eE).

# 3.3 Annual site CH<sub>4</sub> flux budgets

The calculated annual  $CH_4$  budget for the year 2011 of the AU-Wrr-WRR site was -3.83 kg  $CH_4$  ha<sup>-1</sup> yr<sup>-1</sup>. The calculated annual  $CH_4$  budget for the year 2011 of the AU-Wom-WOM site was -1.79 kg  $CH_4$  ha<sup>-1</sup> yr<sup>-1</sup>.

# 4 Discussion

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One of the most novel results of our study is the strong linear relationship observed between soil moisture and CH<sub>4</sub> uptake. To our knowledge the strength of this relationship is unique for temperate forest systems measured using continuous automated chamber systems over a long-period. It is also striking that this strong linear relationship was similar in the two temperate eucalypt forests (dry and wet) regardless of the differences in forest structure, soil type, annual precipitation and geographical distance. It is possible that the two different measurement systems (GC at AU-WRR and FTIR at AU-WOM) could produce different measures of CH<sub>4</sub> flux if operated at the same site because of technological and methodological differences. If that were true, there would only be a remote chance that the two linear relationships between CH4 flux and AFP would overlap one another. As such, our finding that the relationships between CH₄ flux and AFP do converge into one common regression line (as shown in Fig. 4) is worthy of note and suggests similar accuracy between the two measurement systems and similar function in soil CH<sub>4</sub> exchange processes at the two forest sites. CH<sub>4</sub> flux data collected long-term in temperate deciduous forest systems in Europe (Butterbach-Bahl and Papen, 2002) has shown that soil moisture can explain up to 58% of the seasonality in soil  $CH_4$  uptake. Similarly, Kiese et al. (2003) reported that soil moisture could explain up to 53% of the seasonality in CH<sub>4</sub> exchange in a tropical rainforests in Queensland, Australia. Soil moisture influences soil gas diffusivity and is considered the most important factor controlling seasonality of CH<sub>4</sub> uptake in soils worldwide (Dalal et al., 2008;Dalal and Allen, 2008;Smith et al., 2003;Smith et al., 2000;Ball et al., 1997a) and the negative relationship between soil moisture and soil CH₄ uptake reported in this study has been previously reported for other ecosystems (Hartmann et al., 2011; Stiehl-Braun et al., 2011; Castro et al., 1994; Price et al., 2003). This agrees with the theory that soil CH<sub>4</sub> uptake is mainly limited by diffusion in most forest ecosystems (Price et al., 2004) when the sites of microbial CH<sub>4</sub> oxidation are distributed through the surface soil (Stiehl-Braun et al., 2011), and the concentration gradient between soil and atmosphere, which drives the flux, is effectively constant (von Fischer and Hedin, 2007). However, previous field studies have never been able demonstrate so conclusively the strength of the relationship (>90% variation explained) between AFP and soil CH₄ uptake, and for two separate forest systems. To our knowledge the only other study where similarly strong correlations between soil moisture and CH<sub>4</sub> uptake have been reported, was for grassland soils under summer rainfall exclusion (Hartmann et al. (2011).

It is important to note that WFPS has commonly been used to model, or compare, soil CH<sub>4</sub> uptake in different ecosystems (Del Grosso et al., 2000). However, in our study this soil environmental variable was not as effective as AFP in explaining the observed CH<sub>4</sub> flux patterns at the two temperate forest sites. At an individual site level, the relationship between WFPS and CH<sub>4</sub> uptake had the same coefficient of determination as between AFP and CH<sub>4</sub> uptake, however, the slope of the relationship differed between the two forest sites (Fig. 4D). This suggests that WFPS is not the most suitable soil moisture metric to relate soil gas diffusivity to soil CH<sub>4</sub> flux when comparing sites or ecosystems. This is most likely due to the fact that WFPS is a proportional measure that relates VWC to the total soil porosity (equation (4)); compared to AFP that is a direct expression of the air filled pore volume in a given soil (equation (5)).

This demonstrates that soil gas diffusivity is primary related to the volumetric fractions of air (AFP), rather than the volumetric fraction of water in the soil since diffusion through air is much faster than through water (Farquharson and Baldock (2008)).

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Our data also show a very weak influence by soil temperature upon soil CH<sub>4</sub> uptake. This temperature effect appears to be mainly driven by the correlation between soil moisture and soil temperature, which is typical for the climate of the investigated forest systems. After the effect of soil moisture was accounted for soil temperature was only able to account for less than 5% of the remaining variability in soil CH<sub>4</sub> flux at AU-WOM and less than 1.5% of the remaining variability in soil CH<sub>4</sub> flux at AU-WRR. Furthermore, the daily temperature variation in soil CH<sub>4</sub> uptake would have been masked in the analyses because all regression analyses were performed on either chamber cycle or daily uptake means. However, the overall weak but statistically significant temperature dependency of soil CH<sub>4</sub> uptake is unlikely to greatly influence seasonal CH<sub>4</sub> flux variability given that at both sites around 90% of seasonal variability in CH<sub>4</sub> uptake can be explained by soil moisture alone and that soil moisture and temperature are weakly correlated in the investigated forest systems. This was more pronounced at the AU-WOM site because temporal soil moisture variability was greater and we had two years of data compared to one year of data at the AU-WRR site. However, a model that includes soil temperature and soil moisture together performed marginally better based on the AIC as compared to a model that only used soil moisture status in predicting soil CH<sub>4</sub> flux at both of our sites, which is logical based on the fact that all soil microbial processes show a physiological temperature response but it appears that for the MOB temperature response is rather muted at our sites during our measurement timeframe. Furthermore, our data also show that soil CH<sub>4</sub> uptake still continued at a very low WFPS of 10% (VWC = 0.07 g cm<sup>-3</sup>, AFP = 0.59 cm<sup>3</sup> cm<sup>-3</sup>) with CH<sub>4</sub> uptake ranging between -62 to -80  $\mu$ g CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> at this time. We can therefore hypothesize that MOB activity was not severely limited by moisture at the AU-WOM and the AU-WRR sites during the measurement period.

This study reports continuous measurement of soil-atmosphere CH<sub>4</sub> exchange in two temperate eucalypt forests in Australia measured at high temporal resolution for  $\frac{1-2 \text{ years}}{12 \text{ month}}$ . Mean daily CH<sub>4</sub> flux values (AU-Wrr-WRR = - 1.35 to -0.12 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>; AU-Wom-WOM = - 1.36 to -0.11 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) were well within the reported range for other temperate forests in Europe (-2.47 to + 0.26 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>; (Smith et al., 2000)) or worldwide (-10.68 to 0.02 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>; (Dalal et al., 2008;Dalal and Allen, 2008)).

The estimated annual CH<sub>4</sub> uptake of -1.79 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> for AU-Wom-WOM and -3.83 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> for AU-Worr-WRR are comparable to the range of -2.5 to - 3.7 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> reported for temperate beech and spruce forest sites in Germany where CH<sub>4</sub> fluxes were measured with a similar automated system over multiple years (Butterbach-Bahl and Papen, 2002). Globally, a range of -1.31 to -10.5 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> has been reported for temperate forest systems based on short and long-term, automated and manual chamber measurement campaigns (Dalal et al., 2008;Dalal and Allen, 2008). The annual CH<sub>4</sub> uptake rate estimated for AU-Wom-WOM in our study was less than a third of the -5.8 kg CH<sub>4</sub> ha<sup>-1</sup> yr<sup>-1</sup> estimated by Meyer et al. (1997) for soils in the same forest system. This earlier CH<sub>4</sub> sink estimate was based on only five seasonal flux measurements but might also be attributed to the measurements being taken during three dry years (1993 – 1995) when average rainfall was 677 mm yr<sup>-1</sup> (Meyer et al. (1997). In comparison, the years when our study was undertaken (2010 – 2012) the average rainfall was 1063 mm yr<sup>-1</sup>. This may partly explain the greater CH<sub>4</sub> uptake estimate of Meyer et al. (1997) as the lower soil moisture levels may well lead to greater CH<sub>4</sub> uptake rates.

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Given that cool, wet temperate eucalypt forests are often compared to rainforests it is worth noting that the mean annual CH<sub>4</sub> uptake estimated for AU Wrr was similar to that estimated for a tropical rainforest in Queensland, Australia (3.2 kg CH<sub>4</sub> ha<sup>-1</sup> yr +; Kiese et al., 2003). The soils at AU Wrr and the Queensland rainforest also have very similar soil characteristics with regards to pH, bulk density and sand, silt and clay fractions (Kiese et al., 2003; Kiese et al., 2008). However, the mean annual precipitation at the Queensland rainforest site was 2.5 fold greater (4395 mm) than rainfall at the AU Wrr site, which given similar assumed soil drainage properties indicates large geographical differences in the activity, size and/or structure of the MOB on a continental scale.

One of the most novel results of our study is the strong linear relationship observed between soil moisture and CH<sub>4</sub> uptake. To our knowledge the strength of this relationship is unique for temperate forest systems measured using continuous automated chamber systems over a long period. It is also striking that this strong linear relationship was similar in the two temperate eucalypt forests (dry and wet) regardless of the differences in forest structure, soil type, annual precipitation and geographical distance.

CH<sub>4</sub>-flux data collected long term in temperate deciduous forest systems in Europe (Butterbach Bahl and Papen, 2002) has shown that soil moisture can explain up to 58% of the seasonality in soil CH<sub>4</sub>-uptake. Similarly, Kiese et al. (2003) reported that soil moisture could explain up to 53% of the seasonality in CH<sub>4</sub>-exchange in a tropical rainforests in Queensland, Australia. Soil moisture influences soil gas diffusivity and is considered the most important factor controlling seasonality of CH<sub>4</sub>-uptake in soils worldwide (Dalal et al., 2008;Dalal and Allen, 2008;Smith et al., 2003;Smith et al., 2000;Ball et al., 1997a) and the negative relationship between soil moisture and soil CH<sub>4</sub> uptake reported in this study has been previously reported for other ecosystems (Hartmann et al., 2011;Stiehl Braun et al., 2011;Castro et al., 1994;Price et al., 2003). This highlights that soil CH<sub>4</sub>-uptake is mainly diffusion limited in most forest ecosystems (Price et al., 2004) when the sites of microbial CH<sub>4</sub>-oxidation are distributed through the surface soil, and the concentration gradient, which drives the flux (i.e. atmospheric CH<sub>4</sub>-concentration), is effectively constant. However, previous field studies have never been able demonstrate so conclusively the strength of the relationship (>90% variation explained) between AFP and soil CH<sub>4</sub>-uptake, and for two

separate forest systems. To our knowledge the only other study where similarly strong correlations between soil moisturand CH<sub>4</sub> uptake have been reported, was for grassland soils under summer rainfall exclusion (Hartmann et al. (2011).

It is important to note that WFPS has commonly been used to model, or compare, soil CH<sub>4</sub> uptake in different ecosystems (Del Grosso et al., 2000). However, in our study this soil environmental variable was not as effective as AFP in explaining the observed CH<sub>4</sub> flux patterns at the two temperate forest sites. At an individual site level, the relationship between WFPS and CH<sub>4</sub> uptake had the same coefficient of determination as between AFP and CH<sub>4</sub> uptake, however, the slope of the relationship differed between the two forest sites (Fig. 4d). This suggests that WFPS is not the most suitable soil moisture metric to relate soil gas diffusivity to soil CH<sub>4</sub> flux when comparing sites or ecosystems. This is probably because it is a proportional measure relating VWC to the overall soil porosity; whereas AFP indicates the real volume of air filled pores and therefore diffusion capacity (see equation (4) and (5)). This demonstrates that soil gas diffusivity is primary related to the volumetric fractions of air (AFP), rather than the volumetric fraction of water in the soil since diffusion through air is much faster than through water. A similar suggestion was made by Farquharson and Baldock (2008) in relation to models of aerobic nitrification processes in soils.

Our data also show a very weak apparent influence by soil temperature upon soil CH<sub>4</sub> uptake. However, this temperature effect appears to be mainly driven by the correlation between soil moisture and soil temperature, which is typical for the climate for the investigated forest systems. After the effect of soil moisture was accounted for soil temperature was only able to account for less than 5% of the remaining variability in soil CH<sub>4</sub> flux at AU Wom and less than 1.5% of the remaining variability in soil CH<sub>4</sub> flux at AU Wrr. The coefficient of variance for individual chamber cycles (%CV<sub>site</sub>) at both sites was generally greater than the coefficient of variance for the daily averages (%CV<sub>daily</sub>) which demonstrates that spatial variability in soil CH<sub>4</sub> flux within a forest site was greater than daily variability. The daily temperature variation in soil CH<sub>4</sub> uptake will have been masked in the analyses because all regression analyses were performed on either chamber cycle or daily uptake means. However, the weak temperature dependency of soil CH<sub>4</sub> uptake is unlikely to play a defined role in seasonal variability given that around 90% of seasonal variability in CH<sub>4</sub> uptake can be explained by soil moisture alone and that soil moisture and temperature are weakly correlated in the investigated forest systems. This was more pronounced at the AU Wom site because temporal soil moisture variability was greater and we had two years of data compared to one year of data at the AU Wrr site.

Our data also clearly show that MOB activity was not limited within the soil moisture range measured during this study. The increase in CH<sub>4</sub> uptake was linear for a decrease in WFPS over a 20 60% WFPS range, and linear for an increase in AFP over a 0.3 to 0.53 AFP range. Furthermore, our data also show that soil CH<sub>4</sub> uptake still continued at a very low WFPS of 10% (VWC = 0.07 g cm<sup>-3</sup>, AFP = 0.59 cm<sup>-3</sup> cm<sup>-3</sup>) with CH<sub>4</sub> uptake ranging between 62 to  $-80 \mu g$  CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> at this time. We can therefore hypothesize that MOB activity was not limited by moisture at the AU Wom and the AU Wrr sites during the measurement period.

#### **5 Conclusion**

Our field data suggest that the difference in magnitude of CH<sub>4</sub> flux at both sites was based solely on differences in air filled porosityAFP due to site differences in soil bulk density, soil porosity as a near identical relationship between AFP and soil CH<sub>4</sub> uptake existed at both sites. This means that future research should investigate whether simple information about soil bulk density can be used to estimate CH<sub>4</sub> uptake across different eucalypt forest ecosystems in Australia, or in other similar ecosystems globallyThis means that simple information about soil bulk density could be used to estimate CH<sub>4</sub> uptake base rates across different eucalypt forest ecosystems in Australia. Our data further demonstrate that temporal variability in soil CH<sub>4</sub> uptake was predominantly controlled by temporal variability in soil AFP that is linked to soil gas diffusivity. This means that seasonality in CH<sub>4</sub> uptake can be predicted with very high accuracy where information about soil moisture dynamics is available or can be simulated with high certainty. However, since soil texture at both sites was relatively coarse and soils were both clay loams further studies need to establish if the AFP to CH<sub>4</sub> relationship holds true across different soil texture classes. Our results highlight the importance of long-term field measurements in establishing relationships between soil environmental drivers and soil CH<sub>4</sub> uptake and allowing the calibration of models used to calculate global CH<sub>4</sub> sink distribution and magnitude.

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Table 1: Parameters and coefficients of determination (Adj.  $R^2$ ) of linear regression models explaining seasonal variability in mean chamber cycle methane flux (FCH4) at a mixed Eucalyptus obliqua forest stand, Wombat State Forest, Victoria (AU-WomWOM) and at a mixed Eucalyptus obliqua and E. regnans forest stand, Warra LTER between, Tasmania, Australia (AU-WrrWRR). Unstandardised and Satandardised coefficients  $\beta$  (in parentheseis); SD refers to standard deviation of parameter; level of significance as indicated by ANOVA (\*  $\leq$  0.05, \*\*  $\leq$  0.01, \*\*\*  $\leq$  0.001). Predictors:  $T_S$  (Soil temperature), GWC (gravimetric soil water content), AFP (air-filled porosity), and VWC (volumetric soil water content). Presented constants are model intercepts and parameters represent the slopes for the predictor variables.

Dependent Variable	Constant	VWC  (SD =  0.051)	T (CD - 1.09)	AED (CD 0.400)	4 71 72
		7 77 C (SD = 0.031)	$\mathbf{T_S} (\mathrm{SD} = 1.98)$	AFP (SD = 0.488)	$Adj. R^2$
$F_{CH4}(SD = 10.899)$	-92.307***	195.378*** (0.925)	-	-	0.855***
$F_{CH4}(SD = 10.899)$	-19.543***	-	-2.215*** (-0.399)	-	0.158***
$F_{CH4}(SD = 10.899)$	-88.835***	191.664*** (0.907)	-0.254*** (-0.046)	-	0.857***
$F_{CH4}(SD = 10.899)$	53.640***	-	-	-195.378*** (0.925)	0.855***
	Constant	VWC  (SD = 0.055)	$T_{S} (SD = 3.42)$	AFP (SD = 0.402)	$Adj. R^2$
$F_{CH4}(SD = 11.296)$	-75.068***	195.768*** (0.957)	-	-	0.915***
$F_{CH4}(SD = 12.720)$	-6.320***	-	-1.701*** (-0.458)	-	0.209***
$F_{CH4}(SD = 10.607)$	-78.336***	201.671*** (0.982)	0.147*** (0.047)	-	0.900***
$F_{CH4}(SD = 11.296)$	53.943***	-	-	<u>-</u> 195.768*** (0.957)	0.915***
	$F_{CH4}(SD = 10.899)$ $F_{CH4}(SD = 10.899)$ $F_{CH4}(SD = 11.296)$ $F_{CH4}(SD = 12.720)$ $F_{CH4}(SD = 10.607)$	$F_{CH4}(SD = 10.899)$ -88.835*** $F_{CH4}(SD = 10.899)$ 53.640*** $Constant$ $F_{CH4}(SD = 11.296)$ -75.068*** $F_{CH4}(SD = 12.720)$ -6.320*** $F_{CH4}(SD = 10.607)$ -78.336***	$F_{\text{CH4}}(\text{SD} = 10.899) -88.835*** 191.664*** (0.907)$ $F_{\text{CH4}}(\text{SD} = 10.899) 53.640*** -$ $Constant VWC (\text{SD} = 0.055)$ $F_{\text{CH4}}(\text{SD} = 11.296) -75.068*** 195.768*** (0.957)$ $F_{\text{CH4}}(\text{SD} = 12.720) -6.320*** -$ $F_{\text{CH4}}(\text{SD} = 10.607) -78.336*** 201.671*** (0.982)$	$F_{\text{CH4}}(\text{SD} = 10.899) \qquad -88.835^{***} \qquad 191.664^{***} (0.907) \qquad -0.254^{***} (-0.046)$ $F_{\text{CH4}}(\text{SD} = 10.899) \qquad 53.640^{***} \qquad - \qquad \qquad -$ $Constant \qquad VWC \text{ (SD} = 0.055) \qquad T_{\text{S}} \text{ (SD} = 3.42)$ $F_{\text{CH4}}(\text{SD} = 11.296) \qquad -75.068^{***} \qquad 195.768^{***} (0.957) \qquad -$ $F_{\text{CH4}}(\text{SD} = 12.720) \qquad -6.320^{***} \qquad - \qquad \qquad -1.701^{***} (-0.458)$ $F_{\text{CH4}}(\text{SD} = 10.607) \qquad -78.336^{***} \qquad 201.671^{***} (0.982) \qquad 0.147^{***} (0.047)$	$F_{\text{CH4}}(\text{SD} = 10.899) \qquad -88.835^{***} \qquad 191.664^{***} (0.907) \qquad -0.254^{***} (-0.046) \qquad -$ $F_{\text{CH4}}(\text{SD} = 10.899) \qquad 53.640^{***} \qquad - \qquad - \qquad -195.378^{***} (0.925)$ $\hline \textbf{\textit{Constant}} \qquad \textbf{\textit{VWC}}(\text{SD} = 0.055) \qquad \textbf{\textit{T}}_{\textbf{S}}(\text{SD} = 3.42) \qquad \textbf{\textit{AFP}}(\text{SD} = 0.402)$ $\hline F_{\text{CH4}}(\text{SD} = 11.296) \qquad -75.068^{***} \qquad 195.768^{***} (0.957) \qquad - \qquad -$ $F_{\text{CH4}}(\text{SD} = 12.720) \qquad -6.320^{***} \qquad - \qquad -1.701^{***} (-0.458) \qquad -$ $F_{\text{CH4}}(\text{SD} = 10.607) \qquad -78.336^{***} \qquad 201.671^{***} (0.982) \qquad 0.147^{***} (0.047) \qquad -$

Table 2: Parameters and coefficients of determination (Adj. R<sup>2</sup>) of selected linear models in combination with results of a restricted maximum likelihood analysis (REML) explaining seasonal variability in mean chamber cycle methane flux (FCH4) at a mixed *Eucalyptus obliqua* forest stand, Wombat State Forest, Victoria (AU-WOM) and at a mixed *E. obliqua* and *E. regnans* forest stand, Warra LTER between, Tasmania, Australia (AU-WRR). Predictors: T<sub>S</sub> (soil temperature) and AFP (air-filled porosity). REML results: Akaike information criterion (AIC); Estimate of importance for models containing both predictors (*in parentheses*).

Site						
	<b>Dependent Variable</b>	<b>Constant</b>	<u>AFP</u>	$\underline{T}_{\underline{S}}$		
		(Intercept)	(slope)	(slope)	<u>AIC</u>	$Adj. R^2$
<u>AU-WRR</u>	<u>F<sub>CH4</sub></u>	<u>53.640</u>	<u>-195.378</u>	Ξ	<u>5666</u>	0.855
	<u>F<sub>CH4</sub></u>	<u>-19.543</u>	Ξ	<u>-2.215</u>	<u>9657</u>	0.158
	<u>F<sub>CH4</sub></u>	<u>55.587</u>	<u>-193.284 (<b>0.997</b>)</u>	<u>-0.254 (<b>0.003</b>)</u>	<u>5629</u>	0.857
<u>AU-WOM</u>	<u>F<sub>CH4</sub></u>	<u>53.943</u>	<u>-195.768</u>	=	<u>7648</u>	0.915
	<u>F<sub>CH4</sub></u>	<u>-6.320</u>	Ξ	<u>-1.701</u>	<u>13088</u>	0.209
	<u>F<sub>CH4</sub></u>	<u>54.766</u>	<u>-201.671 (<b>0.998</b>)</u>	<u>0.147 (<b>0.002</b>)</u>	<u>7617</u>	0.900

Table 23: Parameters and coefficients of determination (Adj.  $R^2$ ) of linear regression models explaining seasonal variability in mean daily methane flux ( $F_{CH4}$ ) at a mixed *Eucalyptus obliqua* forest stand, Wombat State Forest, Victoria (AU-WomWOM) and at a mixed *Eucalyptus obliqua* and *E. regnans* forest stand, Warra LTER between, Tasmania, Australia (AU-WrrWRR). Unstandardised and standardised coefficients  $\beta$  (in parentheseis); SD refers to standard deviation of parameter; level of significance as indicated by ANOVA ( $^* \leq 0.05$ ,  $^{***} \leq 0.01$ ). Predictors:  $T_S$  (Soil temperature), GWC (gravimetric soil water content), AFP (air-filled porosity), and VWC (volumetric soil water content). Presented constants are model intercepts and parameters represent the slopes for the predictor variables.

(SD = 0.273) (SD = 0.273) (SD = 0.273)	Constant -2.165*** -0.459*** -2.167*** 1.164***	VWC (SD = 0.058)  4.433*** (0.947)  - 4.435*** (0.947)	$T_S \text{ (SD = 2.02)}$ 0.052*** (-0.388) 0.0001 (0.001) -	- 4.433*** (-0.947)	Adj. R <sup>2</sup> 0.896*** 0.148*** 0.895*** 0.896***
(SD = 0.273) (SD = 0.273)	-0.459*** -2.167***	-	-0.052*** (-0.388)		0.148*** 0.895***
(SD = 0.273)	-2.167***	- 4.435*** (0.947) -	· · · · · · · · · · · · · · · · · · ·		0.895***
		4.435*** (0.947)	0.0001 (0.001)	4.433*** (-0.947)	
(SD = 0.273)	1.164***	-	-	4.433*** (-0.947)	0.896***
	Constant	VWC  (SD =  0.055)	$T_S (SD = 3.55)$	AFP  (SD = 0.055)	$Adj. R^2$
(SD = 0.275)	-1.819***	4.771*** (0.962)	-		0.924***
(SD = 0.302)	-0.161***	-	-0.038*** (-0.452)		0.203***
(SD = 0.275)	-1.915***	4.956*** (0.999)	0.004*** (0.053)		0.926***
	1 152***	-	-	-4.771*** (-0.962)	0.924***
(	SD = 0.302) SD = 0.275)	SD = 0.302) -0.161***	SD = 0.302 $-0.161***$ - $SD = 0.275$ $-1.915***$ $4.956*** (0.999)$	SD = 0.302 $-0.161***$ $ -0.038***(-0.452)$ $SD = 0.275)$ $-1.915***$ $4.956***(0.999)$ $0.004***(0.053)$	SD = 0.302) $-0.161***$ $ -0.038*** (-0.452)SD = 0.275$ ) $-1.915***$ $4.956*** (0.999)$ $0.004*** (0.053)$

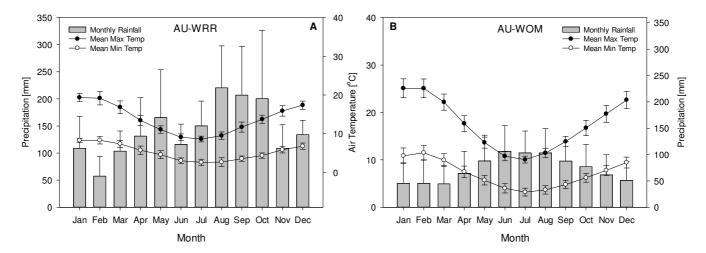


Figure 1: Climate at the investigated sites: Warra LTER in Tasmania (BA, AU-WFFWRR) and Wombat state Forest in Victoria (AB, AU-WomWOM). Closed symbols represent monthly mean maximum air temperatures, open symbols represent monthly mean minimum air temperatures. Bars represent monthly precipitation. Error bars represent ± 1 SESD. Data source Bureau of Meteorology Australia, www.bom.gov.au station numbers 088020 for AU-Wom-WOM and 097024 for AU-WFFWRR.

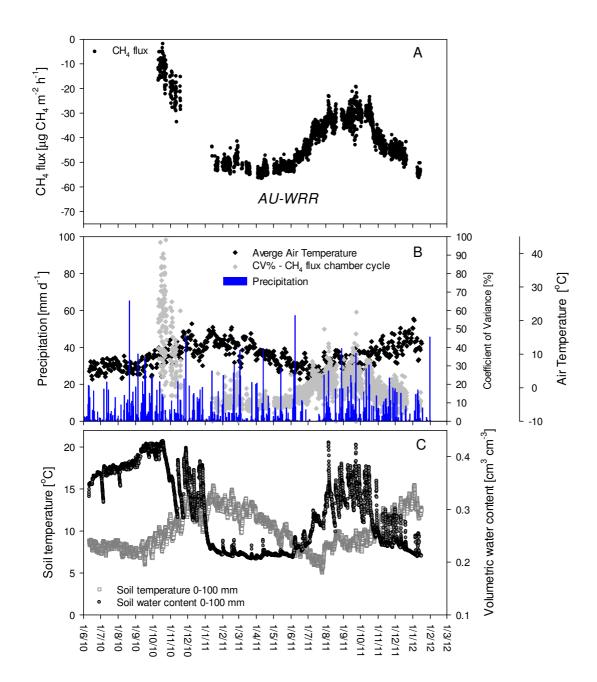


Figure 2: Soil-based flux of CH<sub>4</sub> at a mixed *Eucalyptus obliqua* and *E. regnans* forest stand. Warra LTER, Tasmania, Australia (AU-WrrWRR). Panel A shows CH<sub>4</sub> flux cycle means of ten chambers measured within a four hour time period, Panel A shows CH<sub>4</sub> flux cycle means (10 chambers) per cycle period (4 hours), panel B shows in black closed symbols site air temperature averaged over the chamber cycle period, daily rainfall sums (bars) and coefficient of variance of of the CH<sub>4</sub> flux cycle mean shown in Panel A CH<sub>4</sub> flux for each chamber cycle (grey closed symbols). Panel C shows soil temperature in the top 0-10 cm averaged over each chamber cycle (grey open symbols) and corresponding volumetric soil moisture water content (grey closed symbols) at the site.

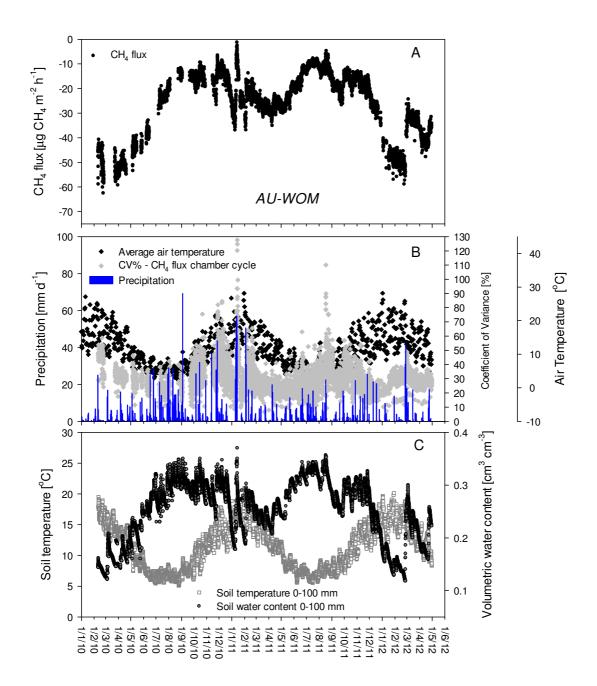


Figure 3: Soil-based flux of CH<sub>4</sub> at a mixed *Eucalyptus obliqua* forest stand, Wombat State Forest, Victoria, Australia (AU-WomWOM). Panel A shows CH<sub>4</sub> flux cycle means of six chambers (10 chambers)measured within a-two hour time periodper cycle period (1.5/2 hours), panel B shows in black closed symbols site air temperature averaged over the chamber cycle period, daily rainfall sums (bars) and coefficient of variance of of the CH<sub>4</sub> flux cycle -mean shown in Panel Afor each chamber cycle (grey closed symbols). Panel C shows soil temperature in the top 0-10 cm averaged over each chamber cycle (grey open symbols) and corresponding volumetric soil moisture water content (grey closed symbols) at the site.

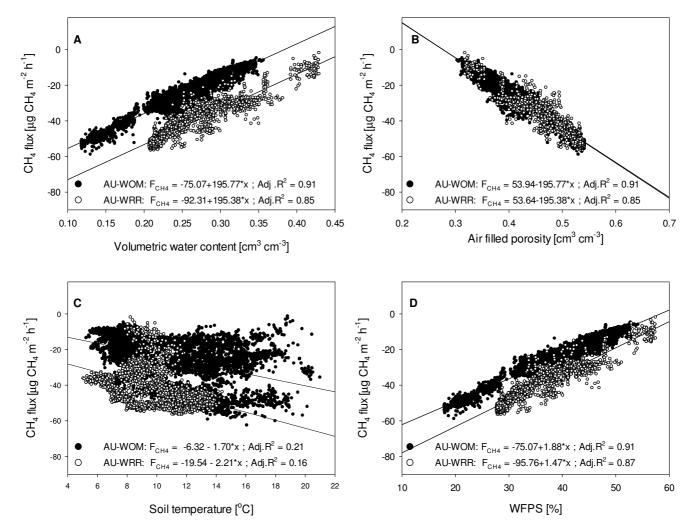


Figure 4: Relationships between soil volumetric water content and soil CH<sub>4</sub> flux (A), soil air-filled porosity and soil CH<sub>4</sub> flux (B), soil temperature and soil CH<sub>4</sub> flux (C) and soil water filled pore space (WFPS) and soil CH<sub>4</sub> flux for each chamber cycle at a mixed Eucalyptus obliqua forest stand, Wombat State Forest, Victoria (closed black symbols, AU-WOM) and at a mixed Eucalyptus obliqua and E. regnans forest stand, Warra LTER between, Tasmania, Australia (open symbols, AU-WRR). Lines (AU-Wom wom = solid line; AU\_Wrr\_WRR = dashed line) symbolise significant linear regressions between the parameters (regression parameters are listed in Table 1).

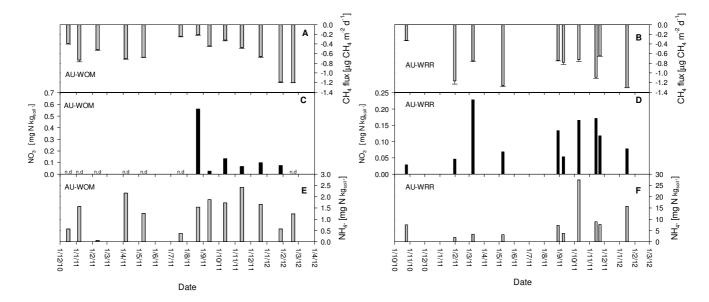


Figure 5: Dynamics in soil CH<sub>4</sub> flux (A, B) soil nitrate levels (C, D) and soil ammonium levels (E, F) at a mixed *Eucalyptus obliqua* forest stand, Wombat State Forest, Victoria (AU-WomWOM) and a mixed *Eucalyptus obliqua* and *E. regnans* forest stand. Warra LTER, Tasmania (AU-WRR), Australia. N.d. = not detectable. Not presented are the results of the linear regression analysis between NH<sub>4</sub><sup>+</sup> and CH<sub>4</sub> flux and NO<sub>3</sub><sup>-</sup> and CH<sub>4</sub> for both sites, these were: AU-WomWOM, NO<sub>3</sub><sup>-</sup>/CH<sub>4</sub> (adj.  $R^2 = 0.06$ , p = 0.21) NH<sub>4</sub><sup>+</sup>/CH<sub>4</sub> (adj.  $R^2 = -0.08$ ); AU-Wrr-WRR NO<sub>3</sub><sup>-</sup>/CH<sub>4</sub> (adj.  $R^2 = -0.11$ , p = 0.80) NH<sub>4</sub><sup>+</sup>/CH<sub>4</sub> (adj.  $R^2 = -0.11$ , p = 0.84).