Dear D.D.R. Owen,

We thank you for your detailed comments on our manuscript and have endeavoured to address your concerns with this reply. All discussion pertaining to Iverach et al. (2015) (unless pertinent to the current manuscript) has not been addressed, as that paper is not in review here. We will address your concerns raised about that manuscript in a future publication as it is a very detailed discussion.

Our sections below align with your point listing.

Section 1. "Addressing a recent paper published in the study area"

We thank you for alerting us to the publication of Owen et al. (2016), published after our submission, and we will certainly reference this work in future iterations of the manuscript.

With regard to "I also encourage the authors to contribute to the conceptual understanding of the system which could be built upon", we believe that our manuscript is making a considerable advance to the conceptualisation of processes occurring in the Condamine Alluvium. Here we provide for the first time microbiological data, which indicate that where CH_4 is detected in the alluvial groundwater it is likely to be sourced from the underlying geological formations.

With respect to your concerns on the usage of isotopic signature, we use the term "signature" in the same context as hundreds of other publications, most notably Whiticar (1999).

Section 2. "Thermogenic vs biogenic CH_4"

In portions of the manuscript we loosely used the term thermogenic when discussing CH_4 within the WCM (and this was also noted by our first reviewer). We will correct this to be in line with the literature. However, this does not change the interpretations in the manuscript.

Section 3. "Limitations on using $\delta^{13}C$-CH_4 as 'signatures'"

We are aware of the limitations of using $\delta^{13}C$-CH_4 to attribute source. Please see our manuscript Zazzeri et al. (2016) doi:10.5194/acp-2016-235, and papers cited in that manuscript.
http://www.atmos-chem-phys-discuss.net/acp-2016-235/. Nonetheless, CH_4 is a useful tracer when interpreted in the context of other hydrogeochemical and microbiological data.

The conclusions reached in this manuscript do not rely on knowledge of the isotopic values of CH_4. There is CH_4 in the alluvium, but our microbiological analyses did not identify any known microbes in the water extracted from the alluvium that would produce CH_4, hence it is sourced elsewhere. We propose in this manuscript that it is sourced from the underlying coal measures. We will leave our isotopic data in the revised manuscripts, because it is useful for readers of the manuscript to have knowledge of these data.

Section 4. "Identifying the $\delta^{13}C$-CH_4 coal measure end-member"

As stated above, identifying the $\delta^{13}C$-CH_4 end-member of the coal, whilst extremely useful, is not critical for assessing the microbiological processes active in the alluvium characterised

in this manuscript for the first time. In our revised manuscript we will refine the papers cited about CH_4 data from the WCM, including referencing Owen et al. (2016).

Section 5. "Microbial activity in the alluvium"

Thank you for drawing our attention to the lack of detail in our sampling protocol. Samples were not filtered in the field, otherwise (as you correctly state) all the biomass would have been left behind on the filter. A complete 16s rRNA sequencing was carried out on the groundwater, fully characterising the bacterial and archaeal gene targets and functional gene targets. No methanogenic archaea were ignored. Culturing experiments would be beneficial for future studies, however they were outside the scope of this research. Microbial data are useful because they tell us what is active within the alluvium. Therefore, if we have CH_4 but no methanogens, or oxidised CH_4 but no methanotrophs, this suggests that the CH_4 is sourced outside of the alluvium and transported upwards to the point of measurement.

As you state, the aquifer is very heterogeneous. We would like to highlight that neither in this manuscript nor in Iverach et al. (2015) did we state that the WCM had a homogeneous $\delta^{13}C$-CH_4 value.

We use the isotopes of DIC and DOC to help us understand what might be occurring in the aquifer. Our hard microbiological data show us that there are no methanogens present, hence there is no methanogenesis occurring in the aquifer. The isotopes of DIC and DOC have simply been employed as a secondary source of data. The fact that they show a trend indicative of no methanogenesis is completely expected, given the absence of methanogens. As you correctly state, the aquifer is heterogeneous and the samples have been taken at different depths, over large intervals – but this just makes it even more interesting that at no location in the aquifer are methanogens found.

You state that an inverse relationship between SO_4^{2-} and CH_4 is typical of methanogenic activity (Owen et al. 2016). However, this does not prove that there is methanogenic activity in the alluvial aquifer. Here we present hard microbiological data that do not support the speculations in Owen et al. (2016).

Section 6. "$\delta^{13}C$-CH_4 are similar to background atmospheric CH_4"

Most of this section is a review of Iverach et al. (2015). Reviewing a published, peer-reviewed paper is not the focus of this discussion forum. We highlight again that the isotopic CH_4 data are not core to the key findings presented in this manuscript. The novel insights into the microbial communities active in the water extracted from the alluvium do not depend upon knowledge of the isotopic composition of the CH_4.

Section 7. Inferred vs. measured $\delta^{13}C$-CH_4 values.

We will modify our wording to stress the difference between measured values and inferred source values.

Section 8. "No samples were taken from the allegedly discharging aquifer".

The focus of our research is on understanding the microbiological communities present in the alluvium. It is not a study on the underlying WCM. Owen et al. (2016) now provide useful

data on the strata underlying the alluvium presenting CH_4 data from both the WCM and the alluvium. Here we show that there are no known methanogens in the alluvium that could produce the CH_4 detected in the alluvial groundwater for the data presented in either Owen et al. (2016) or Iverach et al (2015). It is therefore reasonable to propose that the CH_4 is sourced from the underlying rock strata (including the WCM).

Please note that our discussion focuses on the migration of CH_4, not the movement of water between the Walloon Coal Measures and the Condamine Alluvium.

Section 9. "Incorrect referencing"

Both Kelly and Merrick (2007) and Hillier (2010) provide useful background information to inform the discussions on hydrogeological processes within the Condamine Alluvium. Kelly and Merrick (2007) was in fact peer-reviewed by scientists from various government departments. This report was written for the Cotton Catchment Communities Cooperative Research Centre. The complete series of groundwater knowledge and gaps documents were peer-reviewed at the time of first publication and then again as part of an independent audit. Owen and Cox (2015) reference both these articles.

With respect to Kelly et al. (2014) and Duvert et al. (2015), we will replace these references with Huxley (1982) and Dafny and Silburn (2014).

In regard to "Owen and Cox (2015) did not explicitly assess water movement between the two aquifers" we provide the following quotes from that paper:

"4.1.1. Potential connectivity between the alluvium and CSG groundwater (Cluster A)"

"A number of simple mixing scenarios were performed to test the likelihood that the hydrochemistry of 2005 water sample from well 42231169 was due to the influx of CSG groundwater from the WCM"

"It also indicates that B4 water samples are not indicative of mixing with the underlying WCM groundwater that is typically Na–HCO3–Cl water type, and B4 water types were not observed in bedrock underlying the alluvium in this area."

"In general, no relationships were observed between CSG groundwater in the WCM and the alluvial groundwater."

"as a result recharge processes and alluvium–bedrock connectivity were a focus of this study"

Kind regards,

C.P. Iverach,
on behalf of all authors.