

## ***Interactive comment on “Local artifacts in ice core methane records caused by layered bubble trapping and in-situ production: a multi-site investigation” by R. H. Rhodes et al.***

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This study presents a rich new data set made possible in large part by the technical innovation of continuous-melting of a stick of ice from an ice core with online continuous methane mixing ratio analysis by a laser spectrometer. The results are spectacular, revealing a whole new dimension of trapped gas variation in ice cores. The authors provide a compelling analysis that shows that methane has small but recognizable annual oscillations in mixing ratio, where the annual layers correspond to years in ice age, not gas age, and must arise due to some irregularity of the gas trapping process that has to do with seasonal variations in some physical property of the firn at the firn-ice transition gas trapping level in polar ice sheets. As such these methane oscillations

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have nothing to do with atmospheric variations, but may yield a rich new source of information about the long-standing puzzle of exactly how gases become separated from the atmosphere and recorded in ice cores.

The authors also discuss melt-layer induced methane anomalies but these are not new. Their findings confirm earlier findings that solubility alone cannot explain the observed magnitudes of the anomalies, and rather some other process such as biological methane production by microbes must be occurring, which is a valuable contribution.

Overall, this work will be of interest to a broad spectrum of the ice core community, in particular those concerned with gases and with gas chronologies, the latter being crucial to the broader climate significance of ice core work. Trapped gases in ice cores remain the sole method of reliably reconstructing the past atmosphere and its radiative forcing, making this niche of Earth Science more important than its rather small membership might suggest. This is an excellent piece of science, carefully interpreted and appropriately nuanced. I think it should be published with only minor revisions. I have made some comments below.

An overall comment is that firn air studies have provided a wealth of information about gas trapping mechanisms from an orthogonal (and complementary) perspective to ice core studies, yet they are scarcely mentioned in this manuscript. I would suggest that the authors discuss in their introduction, and include references to, papers that document the existence of a lock-in zone, via the demonstrated ability to pump 1000s of liters of air from this zone without change in trace gas mixing ratios. This implies that a very large amount of air is being withdrawn from an essentially horizontal reservoir, without any leakage from shallower reservoirs. In addition, the measured composition of this air demonstrates in many cases a total absence of halocarbon tracers (Butler et al., Nature 1999; Battle et al. Nature 1996; Severinghaus et al. 2010) The latter paper found the entire Industrial Revolution history of CH<sub>4</sub> and CO<sub>2</sub> contained within only 4 meters of lock-in zone. The only way to explain this observation is that there must be strong horizontal layering and sealing in at least the deeper half of the lock-in zone,

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that is capable of preventing the downward intrusion of younger air. This observation runs counter to some of the concluding remarks in the manuscript.

Butler, J.H., Battle, M., Bender, M.L., Montzka, S.A., Clarke, A.D., Saltzman, E.S., Sucher, C.M., Severinghaus, J.P., and Elkins, J.W., A record of atmospheric halocarbons during the twentieth century from polar firn air, *Nature* 399, 749-755 (1999).

Severinghaus, J.P., M. R. Albert, Z. R. Courville, M. A. Fahnestock, K. Kawamura, S. A. Montzka, J. Mühle, T. A. Scambos, E. Shields, C. A. Shuman, M. Suwa, P. Tans, R. F. Weiss (2010), Deep air convection in the firn at a zero-accumulation site, central Antarctica. *Earth and Planetary Science Letters* 293, 359-367. doi:10.1016/j.epsl.2010.03.003

Specific comments: Pg 4 line 16 This might be a good spot to mention that Etheridge et al suggested that these CH<sub>4</sub> differences represented air-age inversions, in other words, younger air in summer layers underlay older air in winter layers, with a most likely age difference between the two of about 2 years (if my memory serves).

Pg 8 line 5 need a comma between “trapping” and “model”

Pg 10 line 10 would be good to add a reference here, to the statement that methane is more soluble than nitrogen.

Pg 10 line 11 shouldn't this be “lower” rather than “higher”?

Pg 11 line 24 The lock-in depth isn't necessarily the same as the depth at which bubbles first start to form. Generally bubbles start to form 5-10 meters above the lock-in depth. See Buizert et al. (2012). So it is not really sound to infer the lock-in depth from the apparent absence of trapped air. I suggest you cut this sentence, and instead just leave it as you have nicely stated on line 20 that “the data appear to encompass the entire lock-in zone”.

Pg 13 line 9 you might add, for clarity, “. . .minimum estimate of the true signal in the ice.”

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Pg 15 line 10 “the CIC firn air transport model”

Pg 16 line 12 “. . .compensate for differences in the. . .” Pg 19 line 7 “in the ice age domain”

Pg 19 line 17 “the Tunu13 and D4 records. . .”

Pg 19 lines 29-31 This is a misunderstanding of the current state of the Horhold and Freitag work. They did initially say that there was a density crossover, with summer layers being initially lower density and then higher density at the lock-in, but then they retracted that claim after several years. Now they claim that the winter layers are indeed the dense layers at the lock-in zone, as most of us already believed (and indeed as Jakob Schwander and Dave Etheridge wrote many decades ago). This makes more intuitive sense anyway since the impurity loadings are higher in winter. [Of course they are hypothesizing that impurities cause enhanced densification.]

I suggest you put a period after the Gregory reference, then start a new sentence saying something to the effect that the Horhold and Freitag papers suggest that impurities enhance densification and therefore also most likely early bubble close-off.

Pg 23, line 15 The layered bubble trapping process only causes a broadening of the gas age distribution from the point of view of a modeler, who compares the age distributions of a model with empirically-determined diffusivities, before and after adding layered bubble trapping to the model.

In fact, the real-world diffusivities are STRONGLY influenced by the existence of the layers themselves, so this modeling experiment is not realistic. A realistic model experiment would perhaps make the diffusivities be some function of local density. [Then, what you would probably see is that the layering greatly reduces the width of the age distribution in the firn air, which overwhelms the broadening effect of the bubble trapping.]

In any case, the statement that layered bubble trapping broadens the age distribution

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isn't correct when interpreted as a statement about Nature, as most readers will do, rather than the modeling experience under the artificial boundary condition of a fixed and empirical diffusivity. See the last paragraph of Mitchell et al. 2015 for a discussion of the effect of layers on narrowing the age distribution.

Empirical support for this view can be obtained from very-low accumulation sites where layers do not appear to be preserved. Here, the age distribution in the bubbles is much broader than in typical Greenland ice, even after accounting for the accumulation rate differences.

A more quantitative way of saying this is that, in the absence of any layered gas trapping, the age distribution should be set by the time-equivalent thickness of the bubble trapping zone. In practice this time is about 10% of the gas age-ice age difference, independent of accumulation rate.

Yet observed gas age distributions are always much less than predicted by this 10% rule of thumb. The explanation for this seems to be that layers seal the gases off much earlier, creating the well-known "lock-in zone", which prevents younger gases from penetrating down into the full thickness of the bubble closure region. The lock-in zone is not perfect; some downward diffusion of gases does occur with respect to the ice, as shown by anthropogenic tracers in firn air studies (Buizert et al 2012 ACP). But this is a second-order effect.

The origin of the CH<sub>4</sub> oscillations is probably in that part of the bubble closure zone that extends ABOVE the lock-in zone. Here, younger air can easily diffuse down and be captured in summer layers that remain open just above the lock-in horizon. This is a testable hypothesis and you might consider mentioning it.

In summary, I would advise a re-wording along the lines of, "Layered bubble trapping has the effect of broadening the modeled age distribution of the air in ice cores, relative to a model scenario without layered bubble trapping but the same prescribed firn air diffusivity profile. Age distributions in realistic models of non-layered firn vs. layered

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firn that capture the effect of layering on the diffusivity have yet to be studied, to our knowledge."

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