The ice-core record of atmospheric methane:

chemistry-climate interactions on tens to thousands of years

Levine, J. G.[1], E. W. Wolff[1], A. E. Jones[1], M. A. Hutterli[1], L. C. Sime[1], O. Wild[2],

P. J. Valdes[3], P. O. Hopcroft[3], A. T. Archibald[4,5], G. D. Carver[4,5], N. J. Warwick[4,5], and J. A. Pyle[4,5]

1 British Antarctic Survey NATURAL ENVIRONMENT RESEARCH COUNCIL

2 LANCASTER UNIVERSITY

3 University of BRISTOL

4 UNIVERSITY OF CAMBRIDGE

5 National Centre for Atmospheric Science NATURAL ENVIRONMENT RESEARCH COUNCIL

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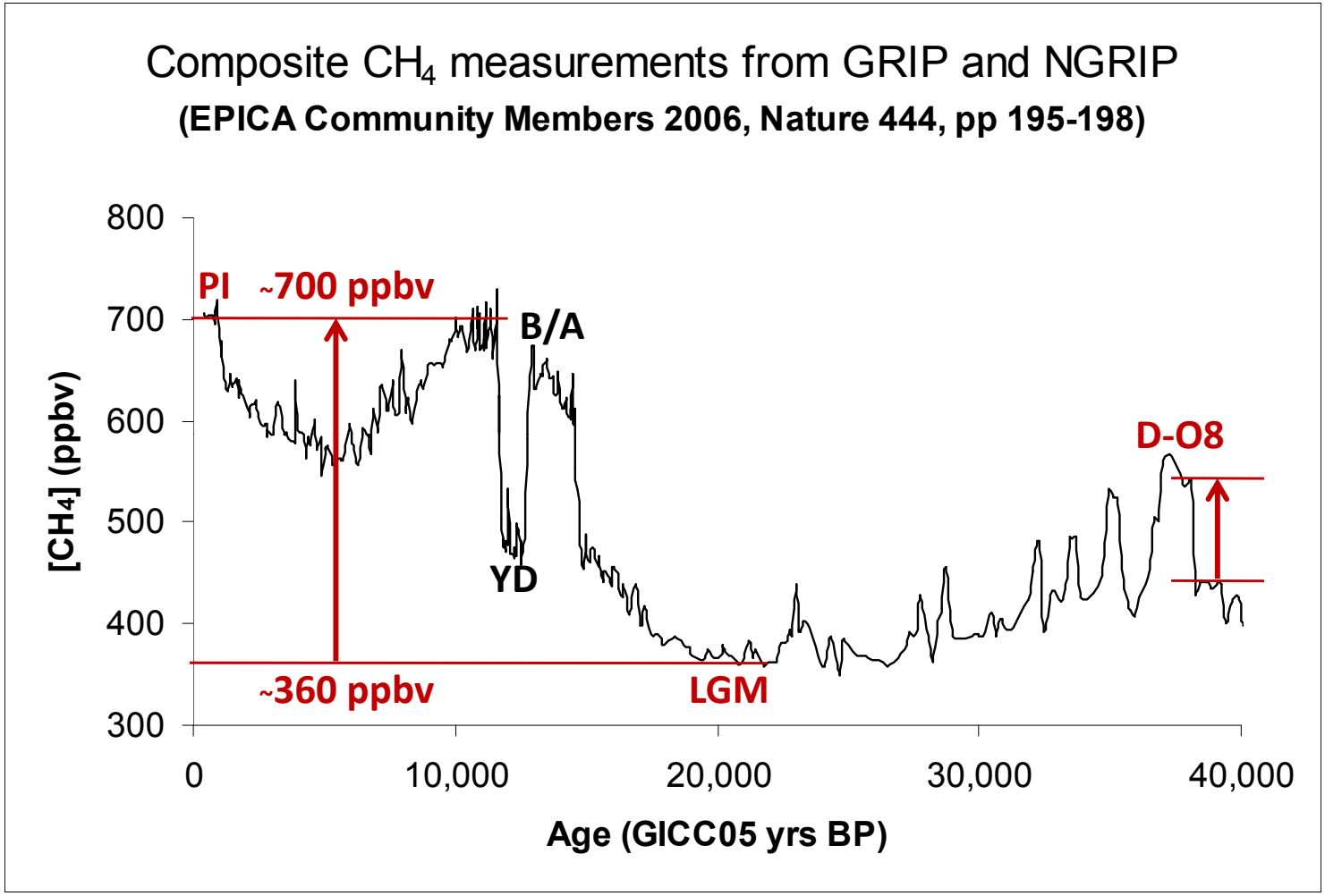
The ice-core record of atmospheric methane



[CH_4] almost doubled between the Last Glacial Maximum (LGM) and the pre-industrial era (PI),

and rose rapidly by 100-200 ppbv at the beginning of each Dansgaard-Oeschger event (e.g. D-O8)

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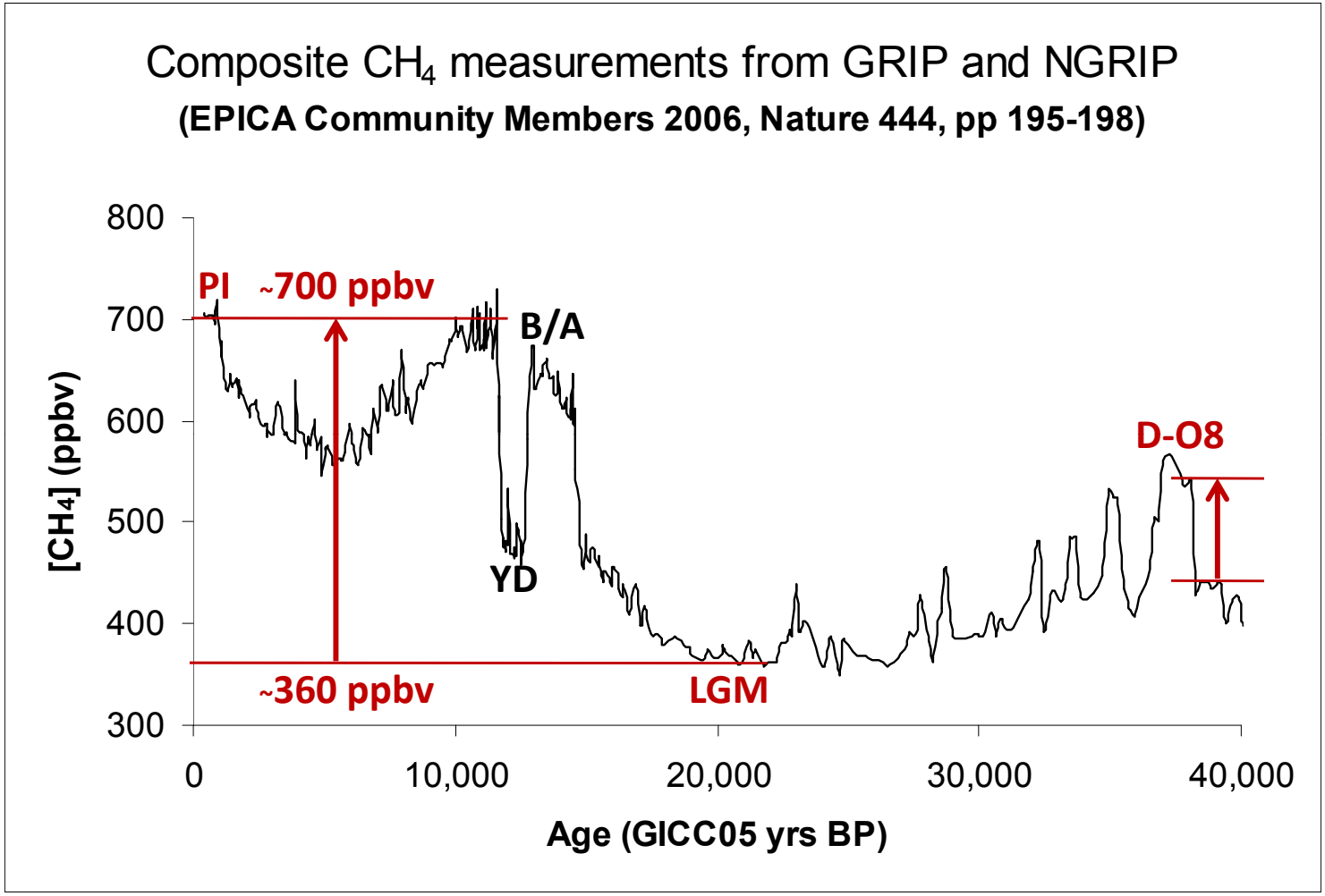

Model study 1: Sources & Sinks – Exploring the drivers of the LGM-PI change in [CH_4]

Model study 2: Isotopes – Exploring influences on $\delta^{13}CH_4$ (a complementary constraint)

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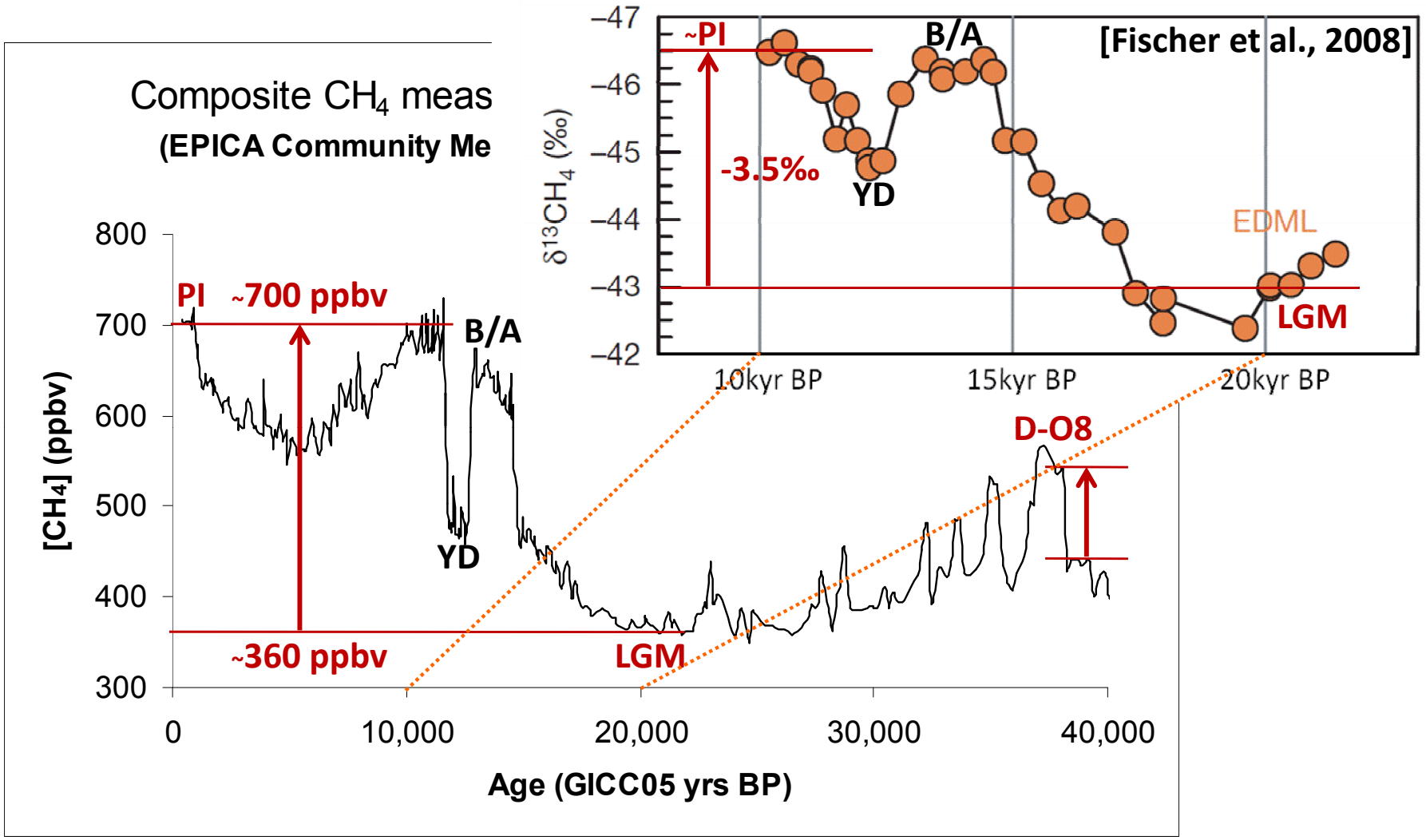


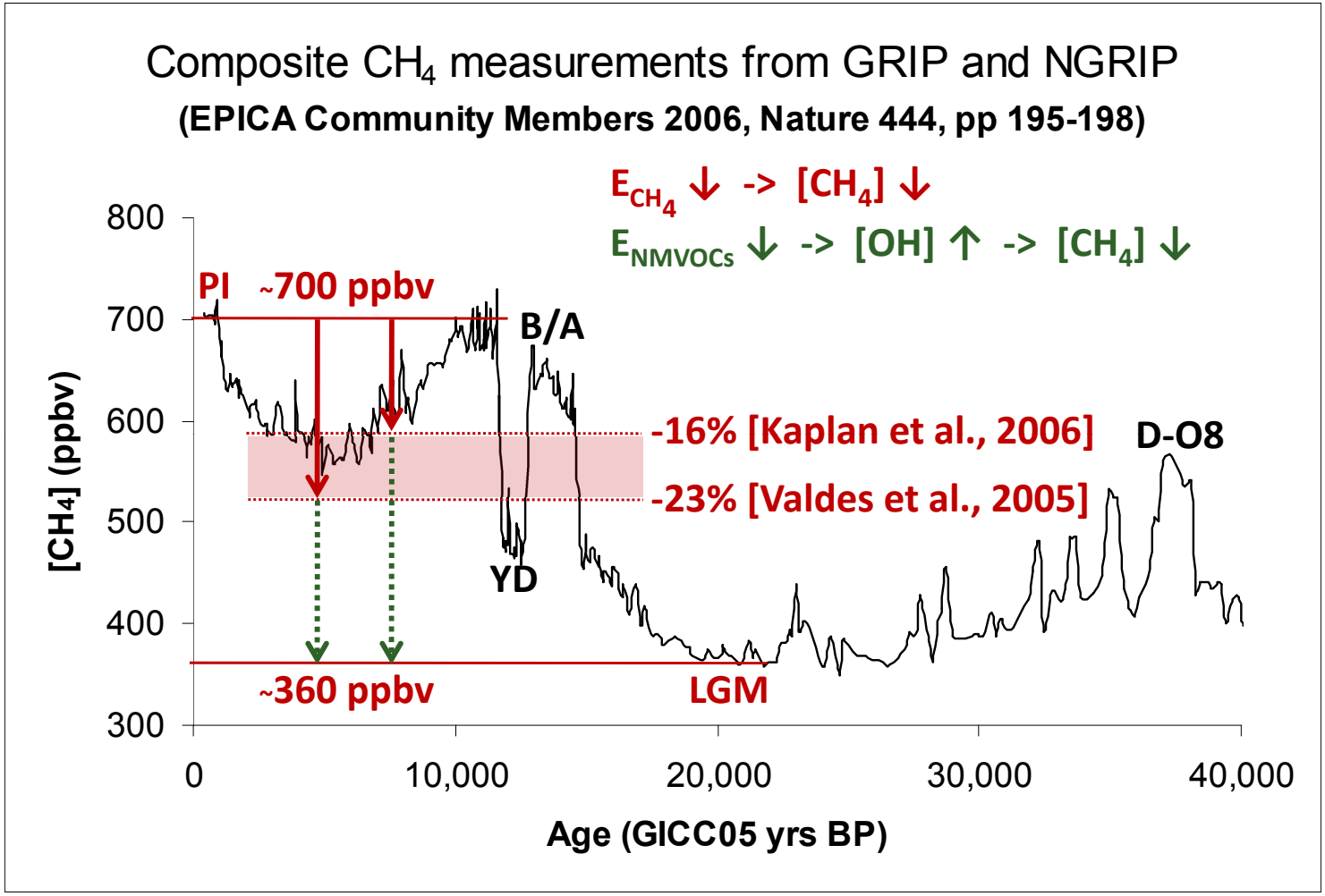
Model study 1: Sources & Sinks – Exploring the drivers of the LGM-PI change in [CH_4]

Model study 2: Isotopes – Exploring influences on $\delta^{13}CH_4$ (a complementary constraint)

Model study 1: Sources & Sinks

Can we reconcile recent bottom-up model estimates of the changes in CH_4 sources and sinks between the LGM and the PI with the observed change in [CH_4]?

Model study 1: Sources & Sinks



Until recently, bottom-up model studies attributed less than half the reduction in $[CH_4]$ to changes in CH_4 sources, appealing to an increase in [OH] (due to reduced NMVOC emissions) to explain the remainder

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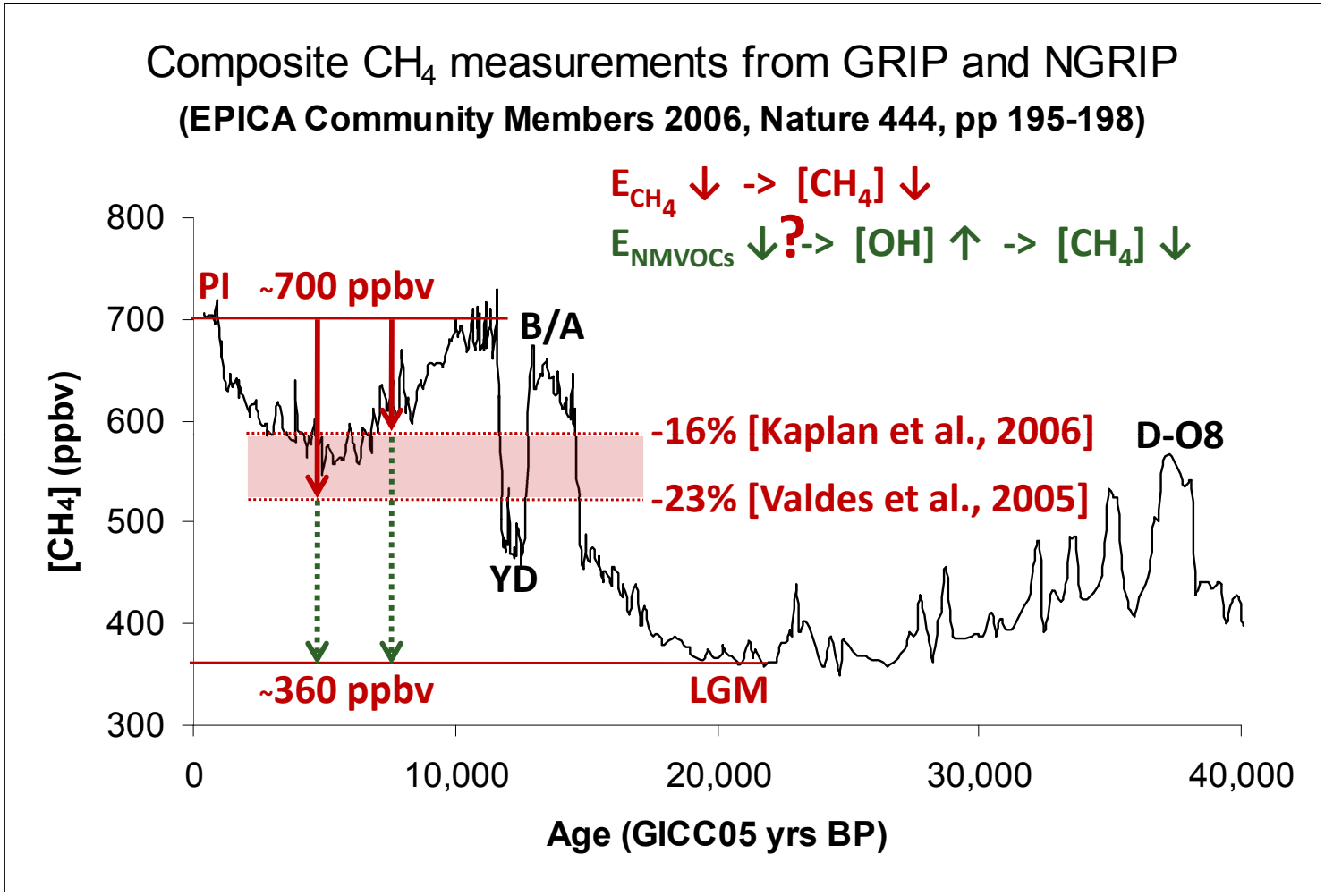

But some OH 'consumed' in isoprene oxidation may be recycled [e.g. Lelieveld et al., 2008],

and changes in humidities and chemical kinetics at the LGM would have tended to <u>increase</u> $[CH_4]$

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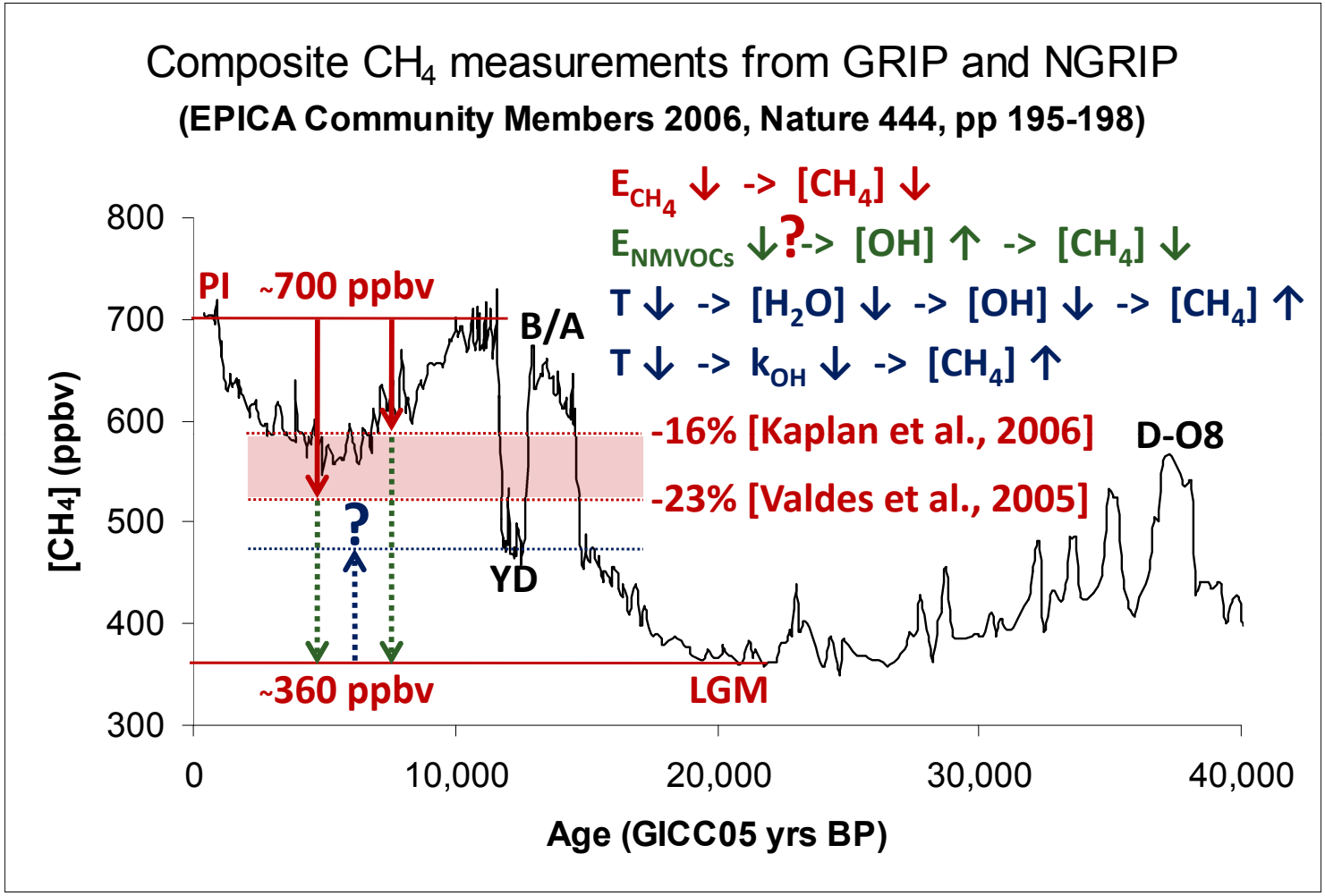

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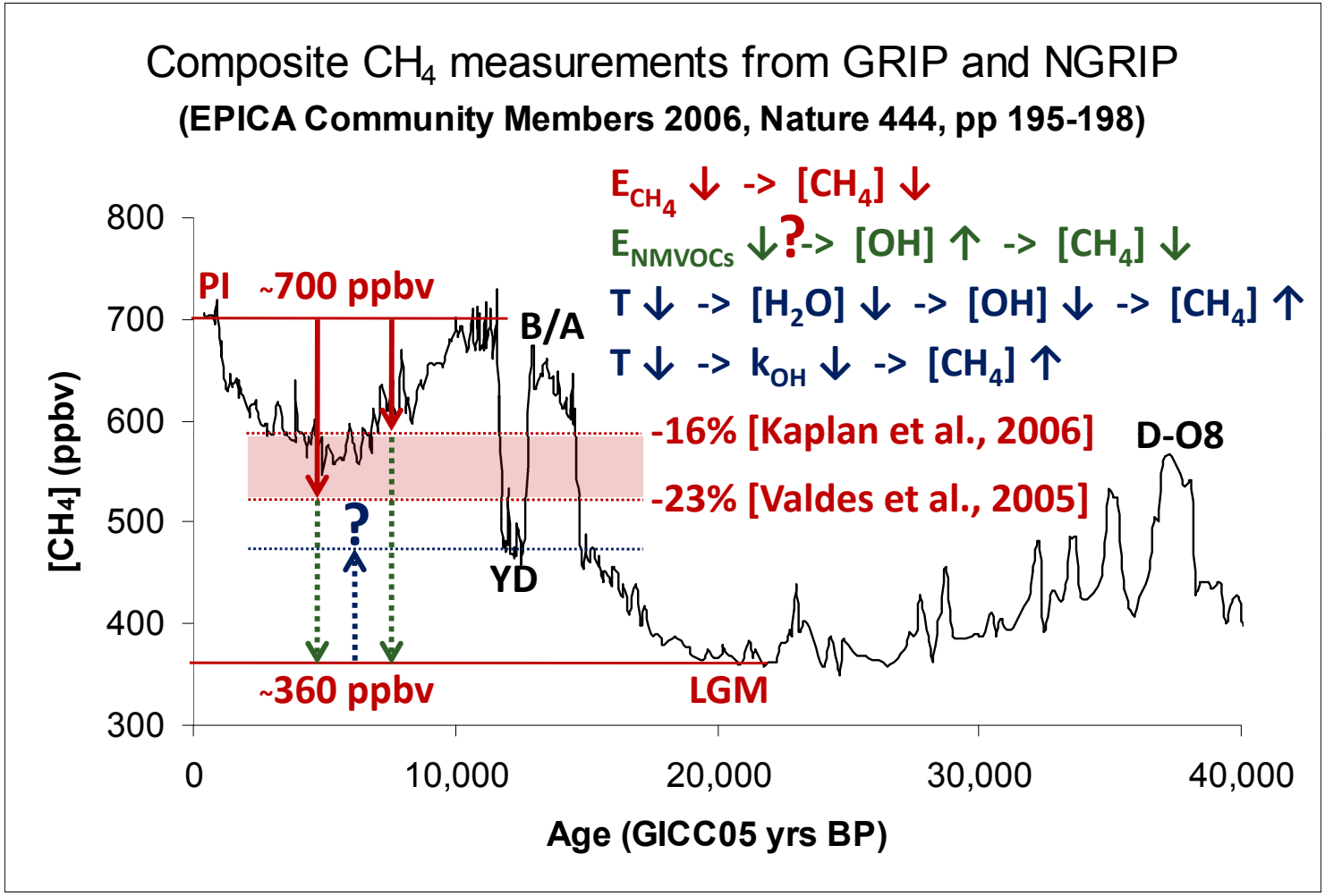

Meanwhile, Weber et al. [2010] have published a higher estimate of the reduction in CH_4 emissions from wetlands at the LGM: 35-42% c.f. 27% [Valdes et al., 2005] and ~0% [Kaplan et al., 2006]

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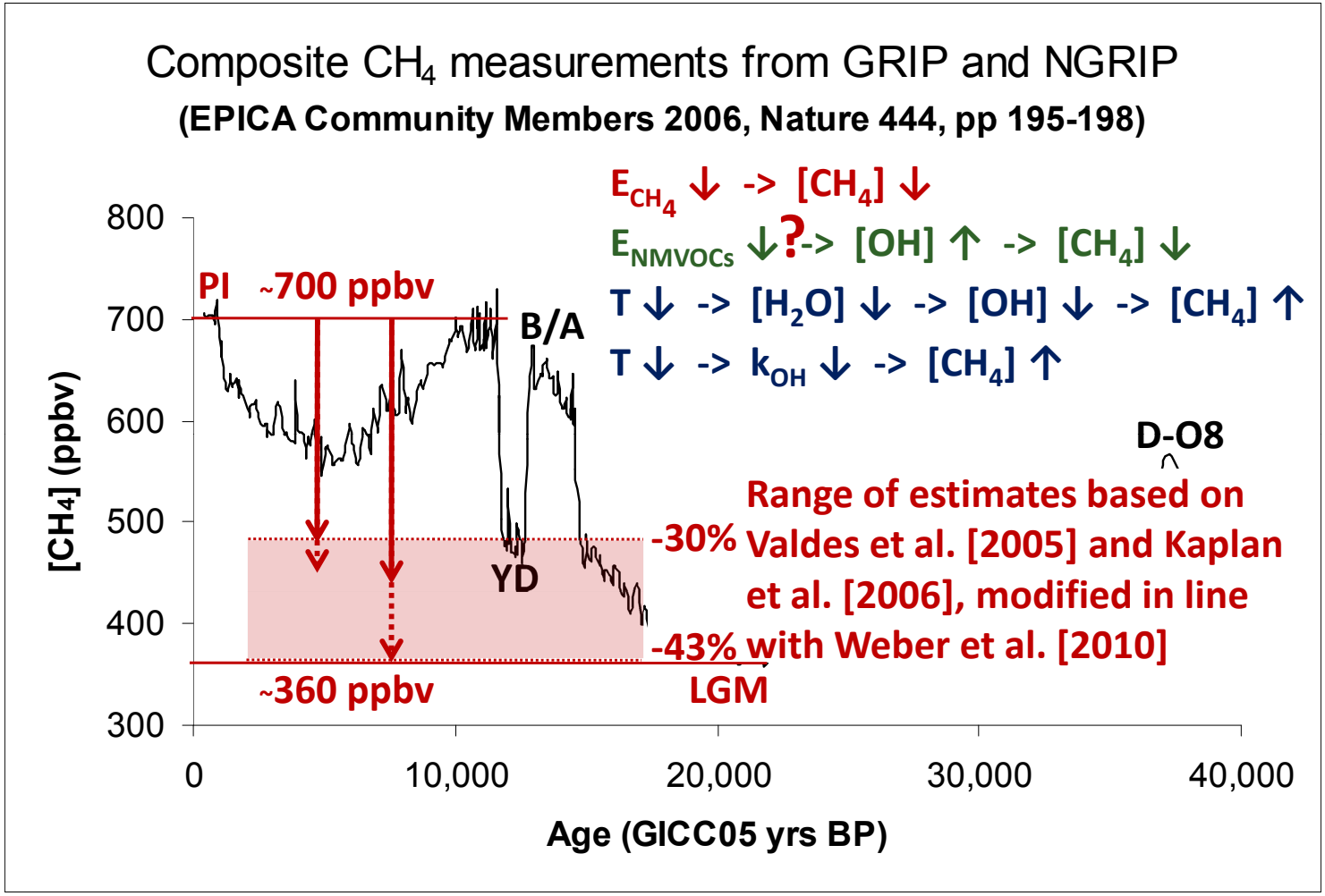

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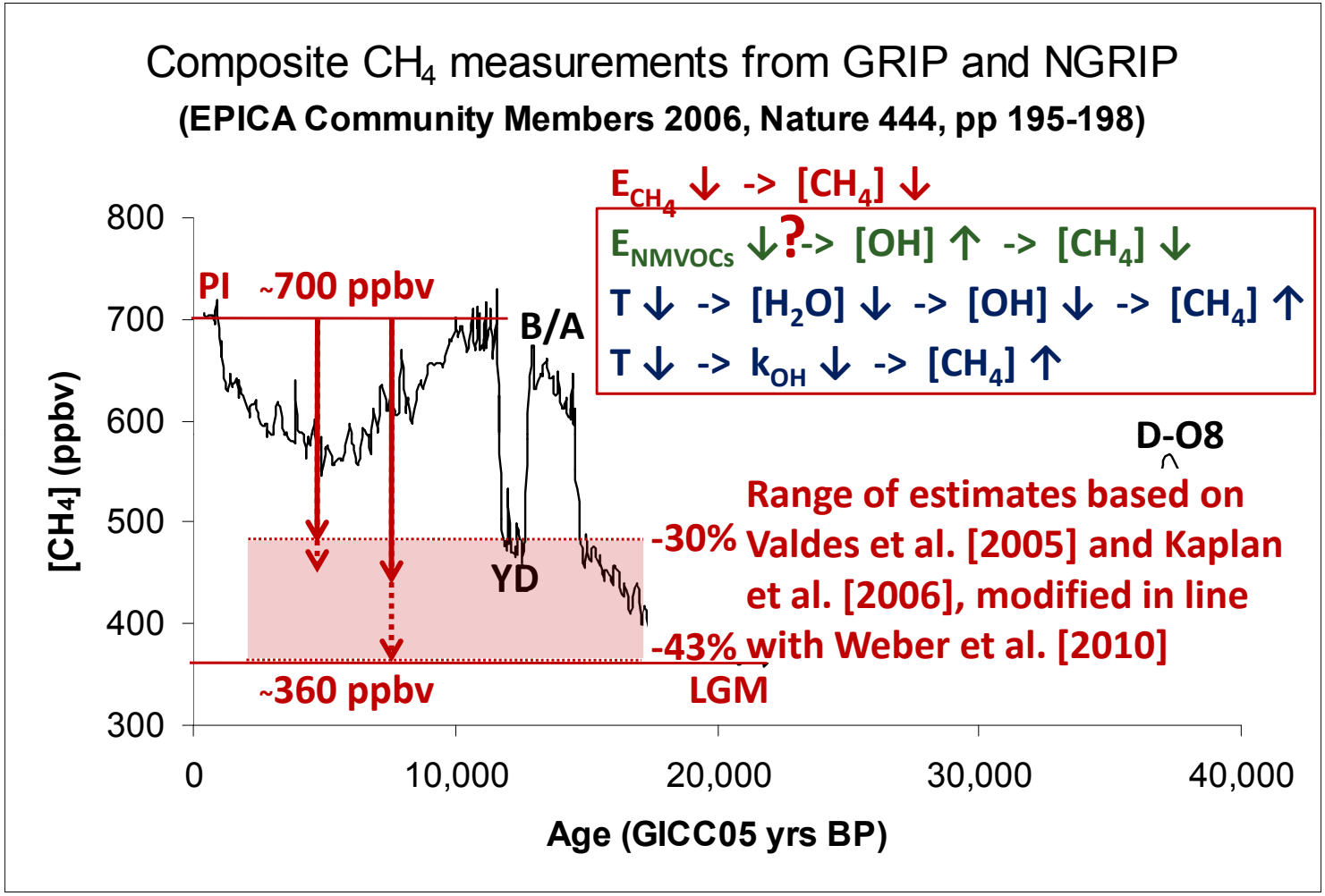

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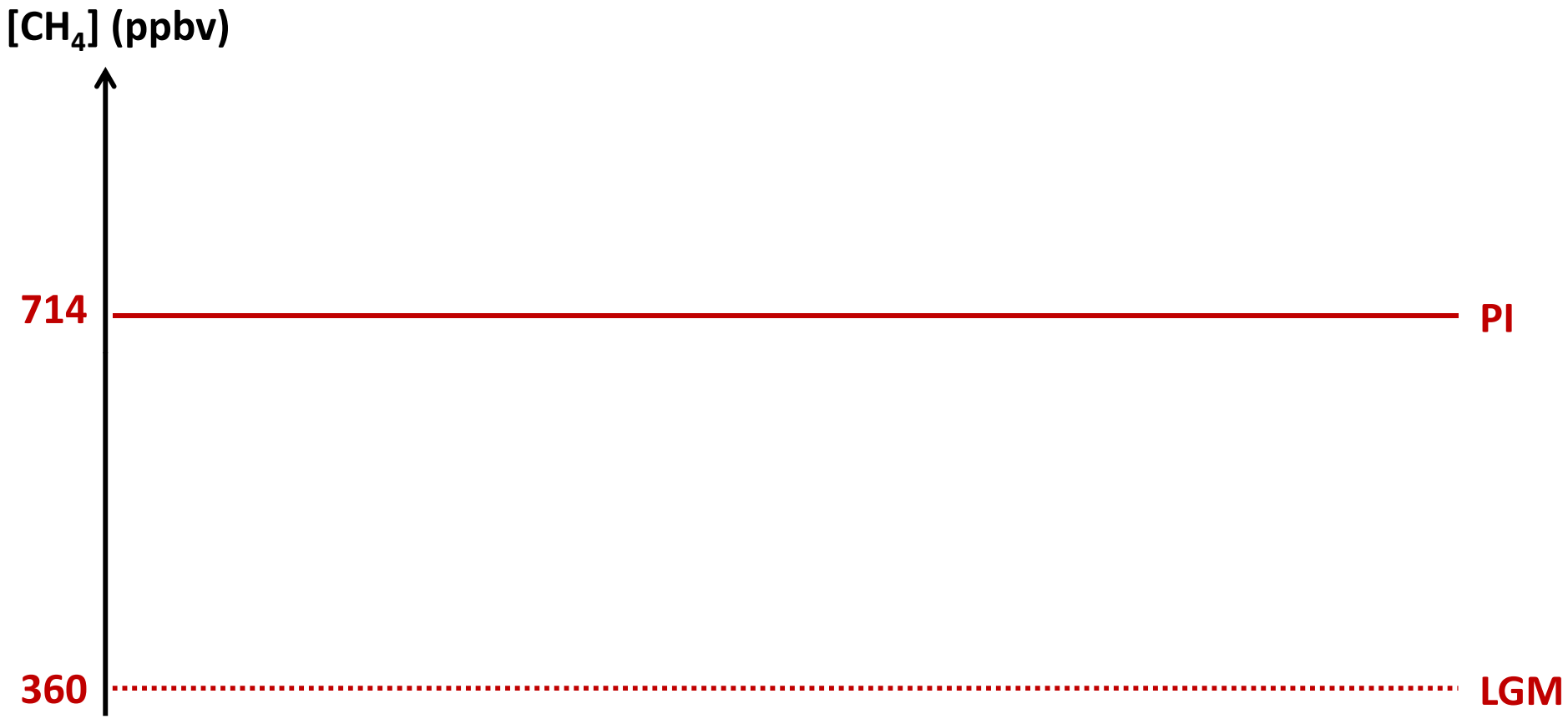

Sensitivity experiments with Cambridge p-TOMCAT model (3D global atmospheric chemistry-transport model) to explore the effects of changes in NMVOC emissions and air temperatures

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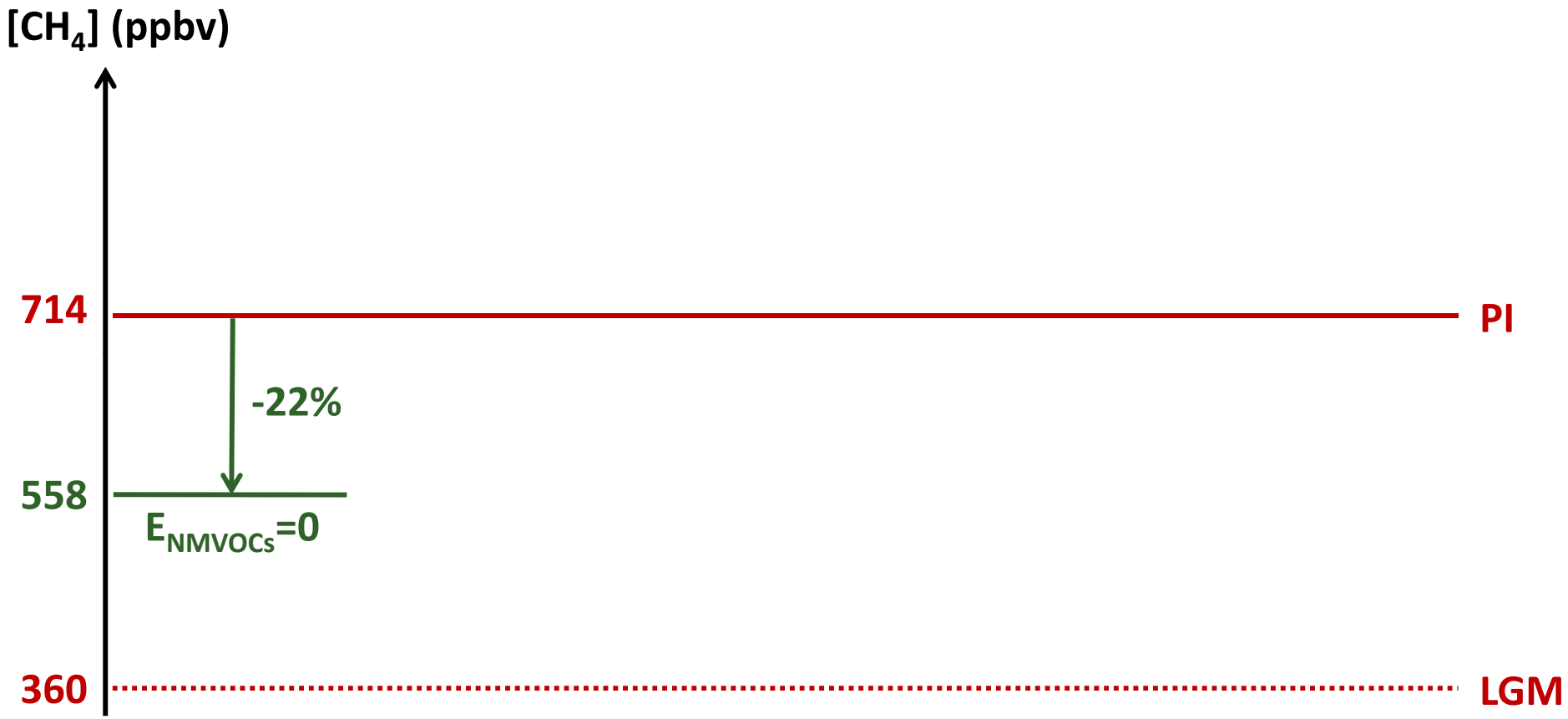

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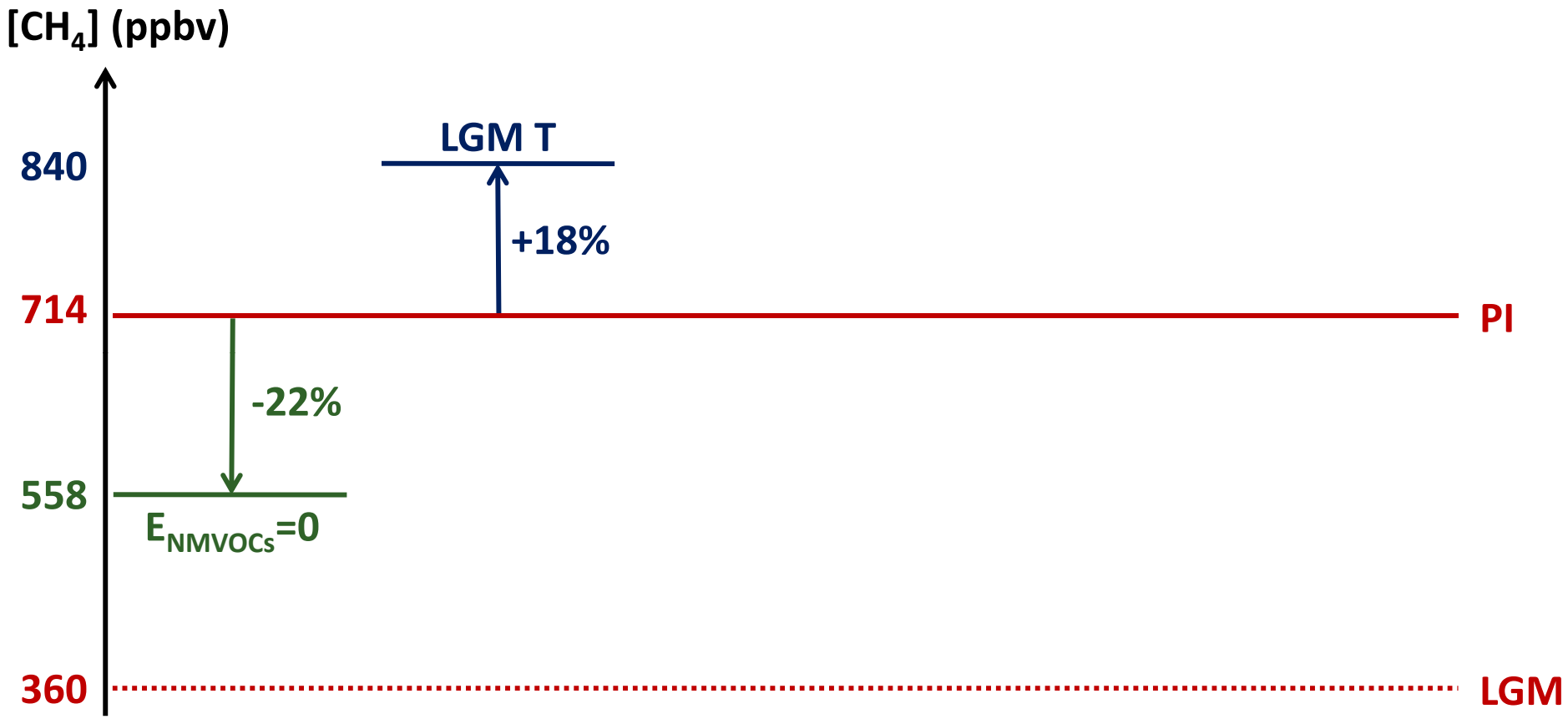

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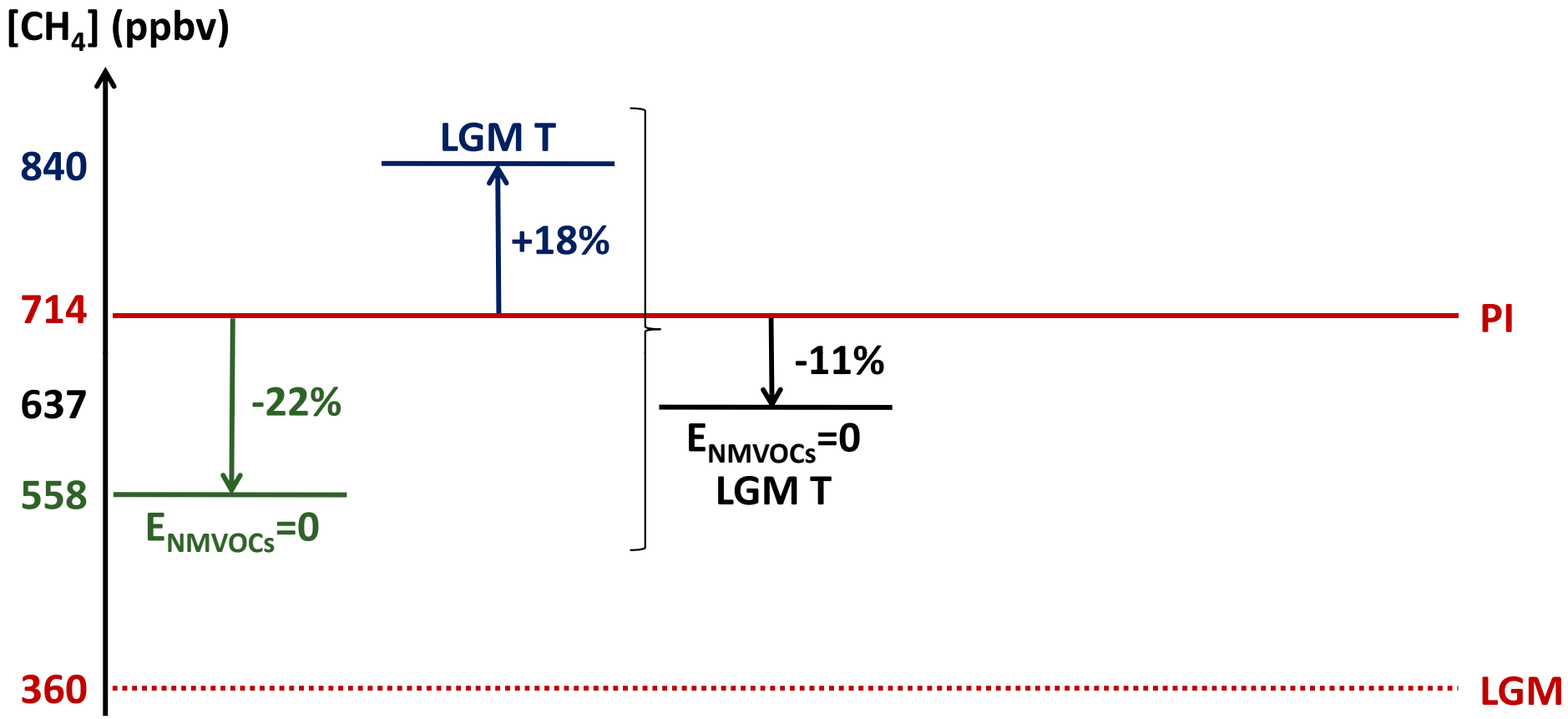

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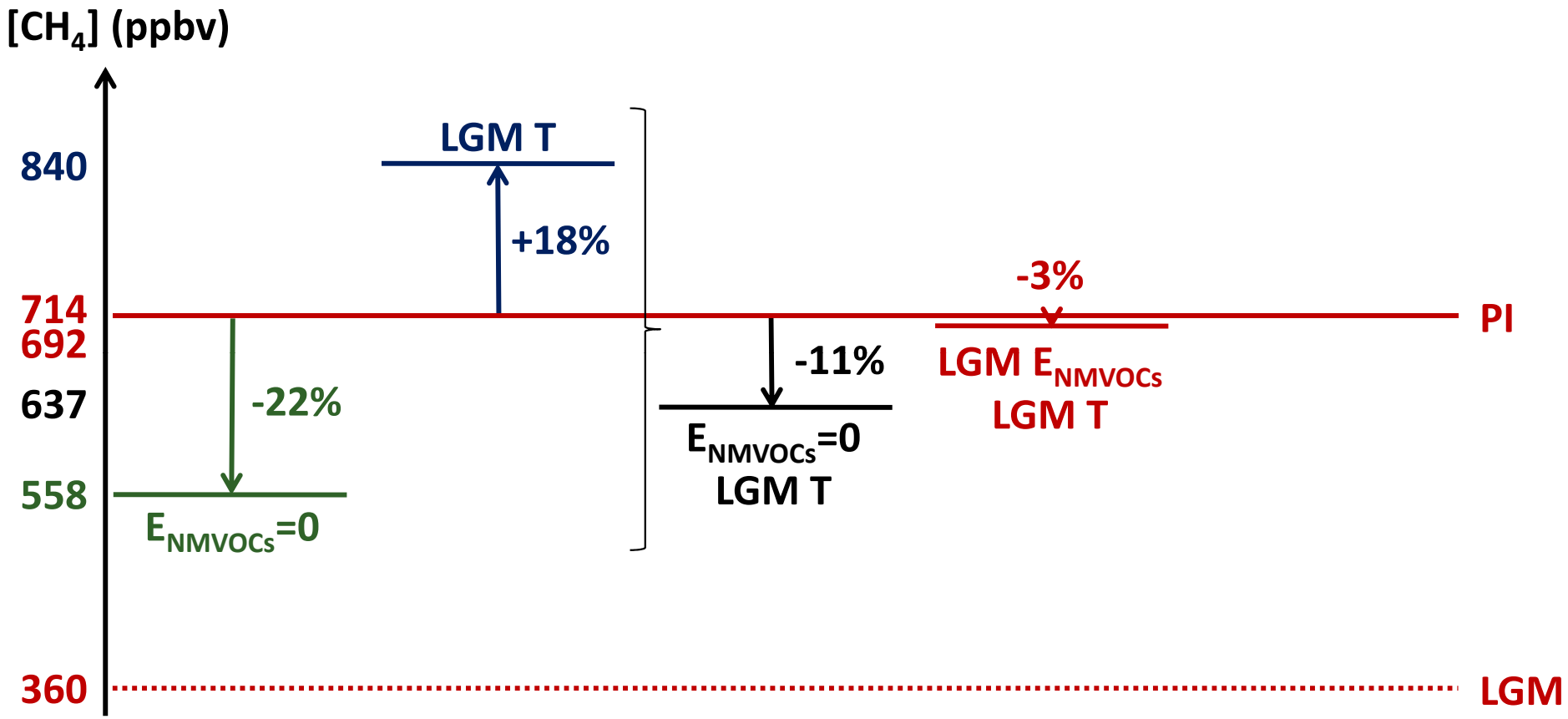

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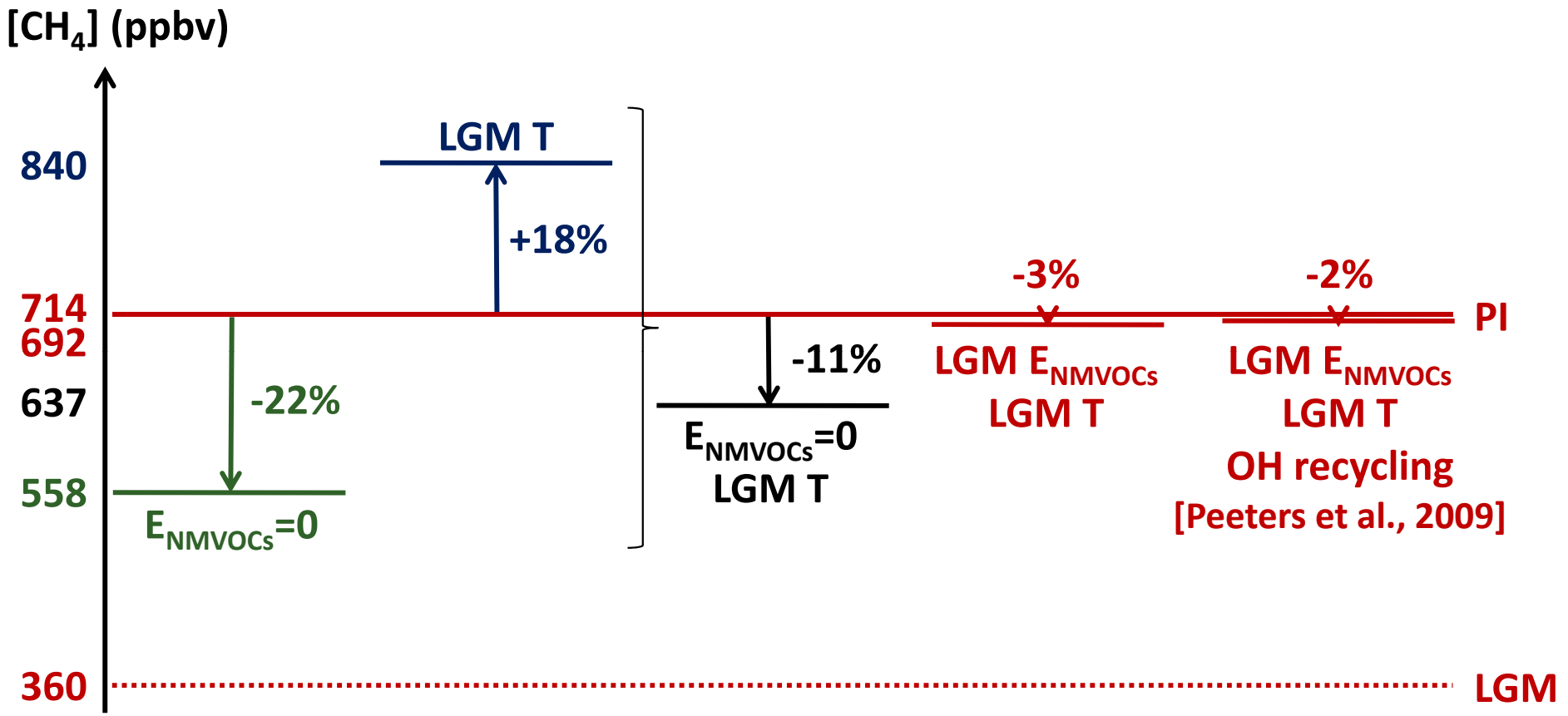

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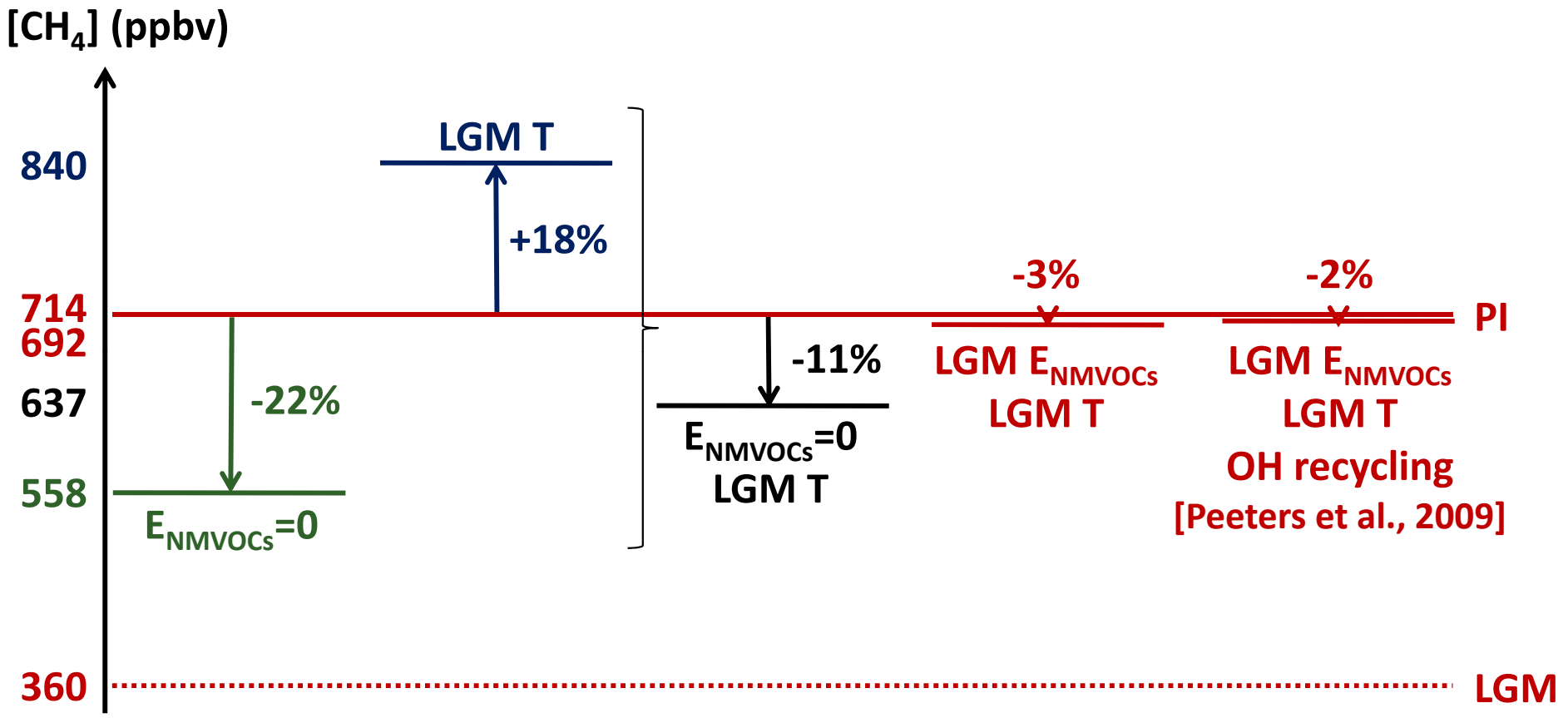

If the net effect of the changes in E_{NMVOCs} and T was negligible, the LGM-PI change in $[CH_4]$ must have been almost entirely source-driven (provided no other factor significantly altered the oxidising capacity)

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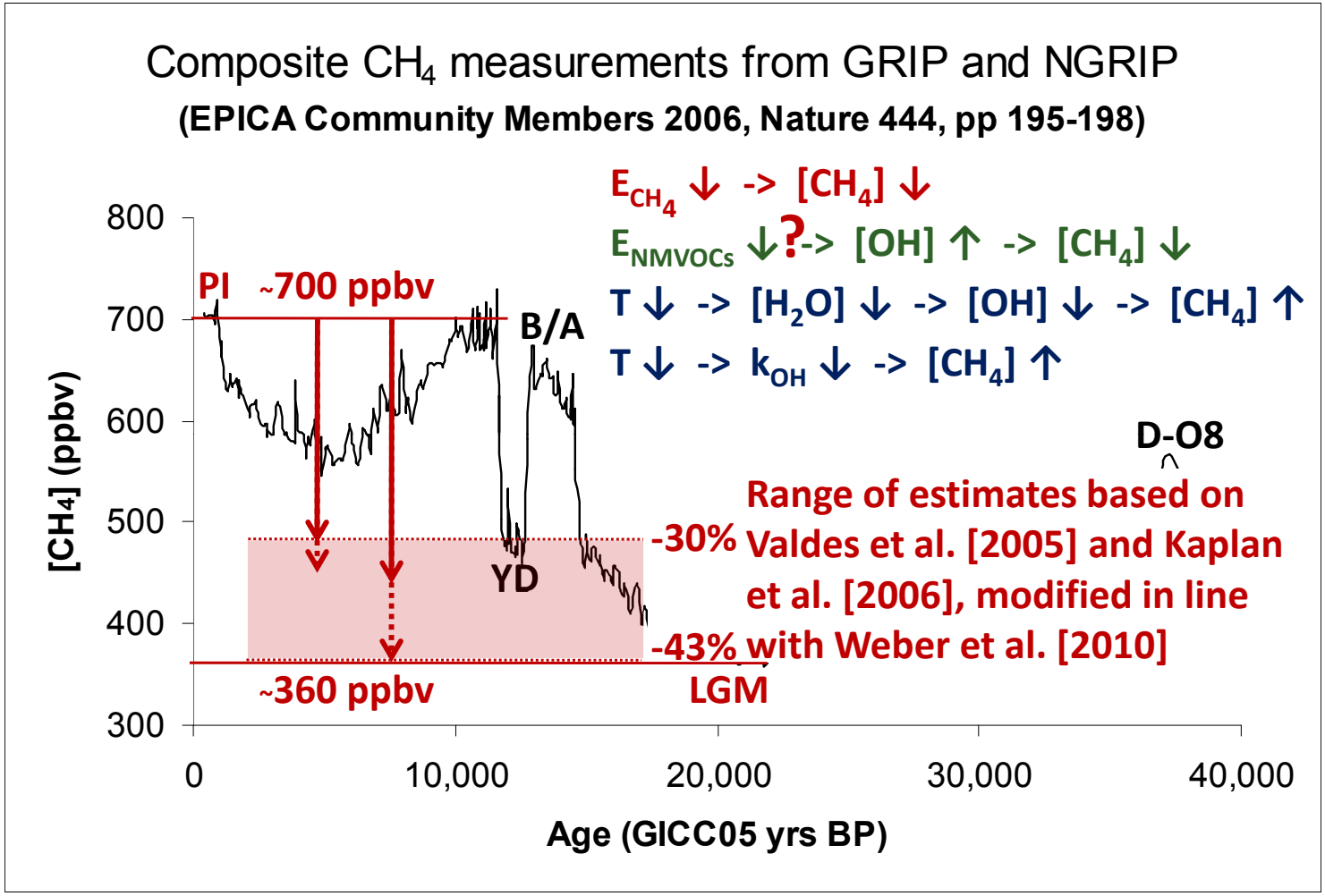

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Model study 1: Sources & Sinks

It is plausible the LGM-PI change in [CH_4] was entirely source-driven, and we can thereby reconcile recent bottom-up model estimates of the changes in CH_4 sources and sinks

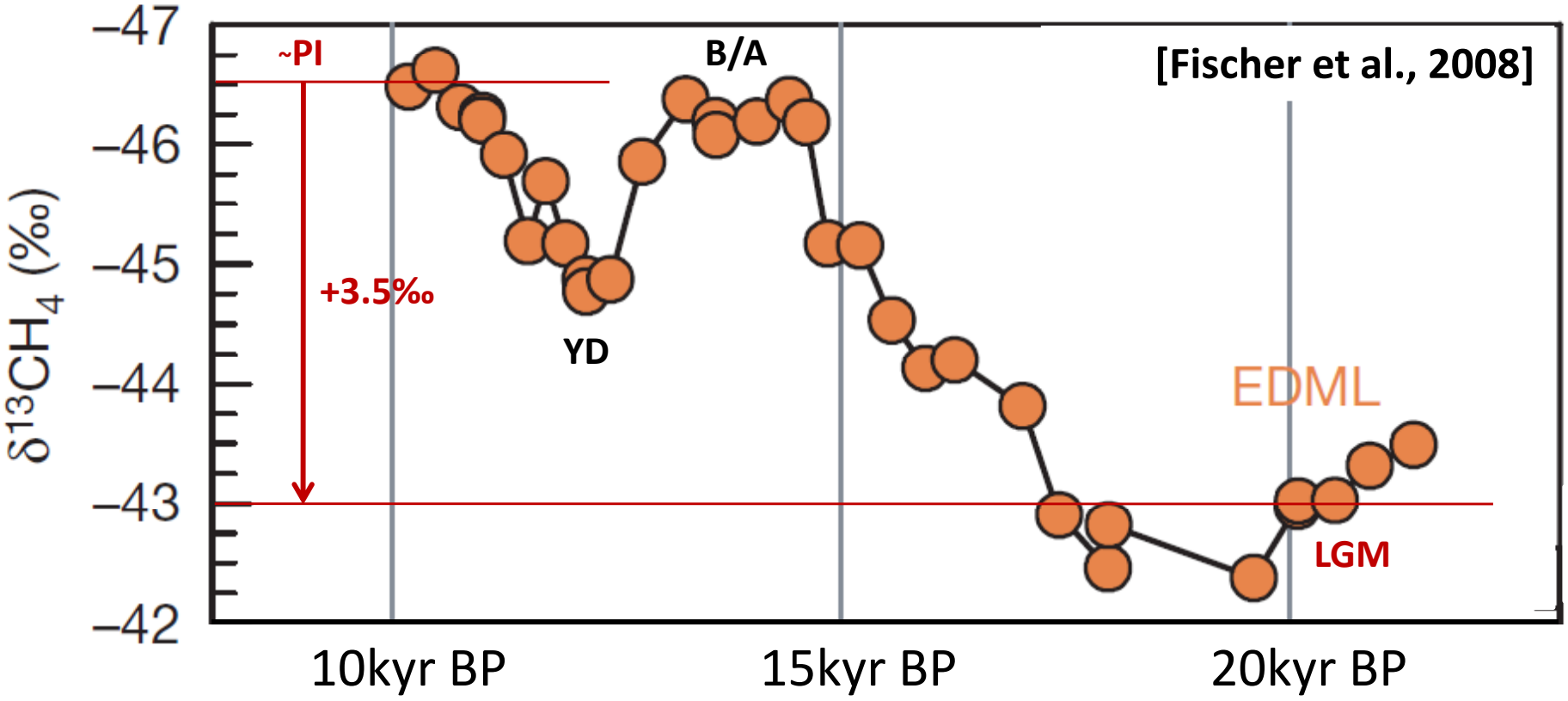
'Reconciling the changes in atmospheric methane sources and sinks between the Last Glacial Maximum and the pre-industrial era' [Levine et al., 2011a, GRL]

An extension of this work to an idealised D-O event [Hopcroft et al., 2011] suggests the rapid rises in [CH_4] at the beginning of D-O events were also essentially source-driven [Levine et al., submitted]

Model study 2: Isotopes

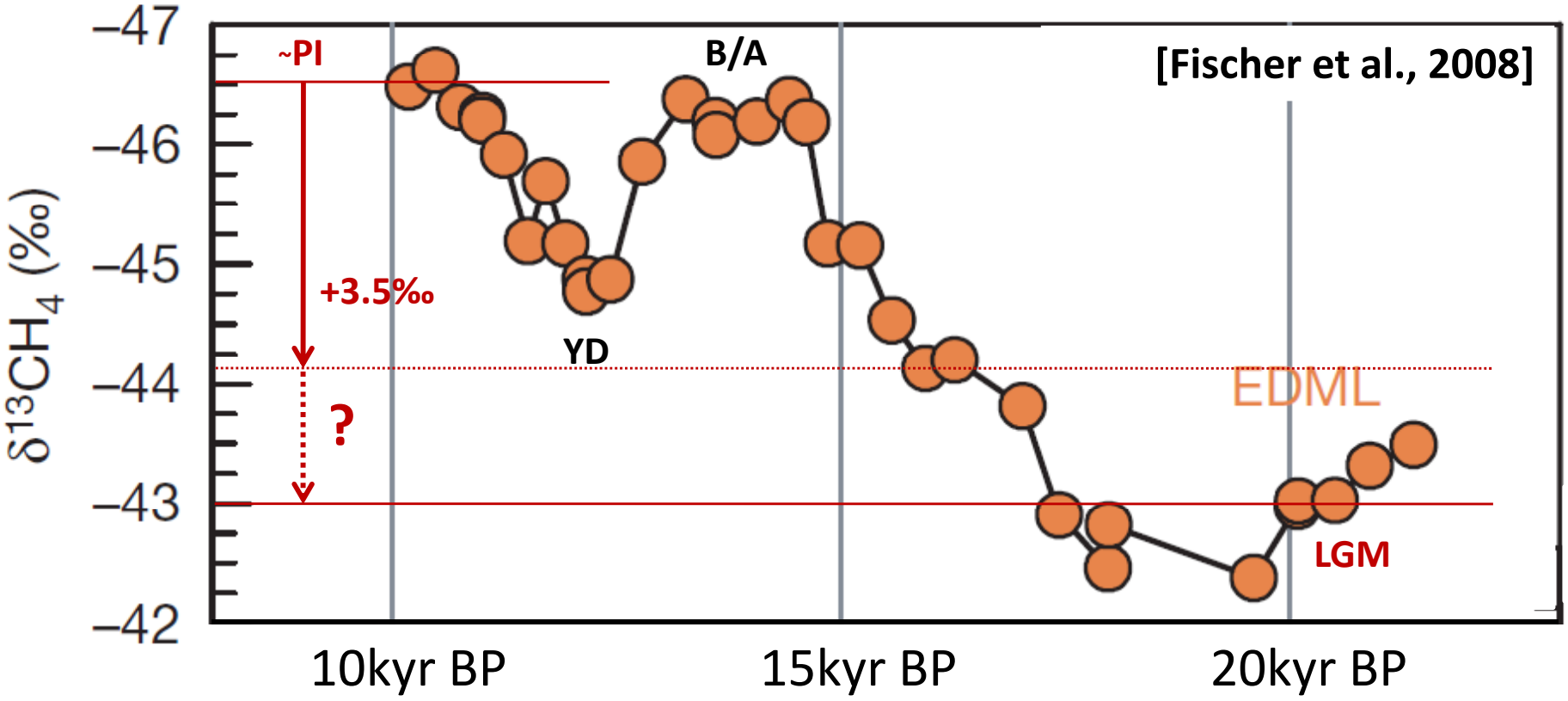
Could changes in the strength of a minor CH_4 sink—oxidation by atomic chlorine in the marine boundary layer (Cl_{MBL})—have contributed to glacial-interglacial changes in $\delta^{13}CH_4$?

Model study 2: Isotopes



Fischer et al. [2008] attributed the 3.5‰ enrichment in $\delta^{13}CH_4$ at the LGM to a near-shutdown of boreal wetland sources of ^{13}C-poor CH_4 accompanied by not dissimilar biomass burning sources of ^{13}C-rich CH_4

Model study 2: Isotopes



But the few charcoal records that span the LGM suggest there was less biomass burning during the last glacial period [Power et al., 2008], so we may still have some enrichment in $\delta^{13}CH_4$ at the LGM to explain

Model study 2: Isotopes

Meanwhile, changes in the strength of the Cl_{MBL} sink have been invoked to explain recent spatial and inter-annual variations in $\delta^{13}CH_4$ [Allan et al., 2005, 2007]

Cl_{MBL} is derived from sea salt aerosol (SSA) [e.g. Vogt et al., 1996], the production of which is highly sensitive to the wind speed at the sea surface [e.g. Monahan et al., 1986]

Polar-ice records show large increases in dust and sea salt at the LGM, relative to the PI, that could be indicative of changes in the atmospheric circulation [e.g. Fischer et al., 2007]

Calculations combining Allan et al.'s [2007] formulation for [Cl_{MBL}], Gong et al.'s [2002] wind-speed dependence of SSA production; and winds (and temperatures) from GCM simulations

Model study 2: Isotopes

We calculate changes in $\delta^{13}CH_4$ at the LGM relative to the PI, driven solely by changes in the circulation, of the order of 10% of the glacial-interglacial difference, from -0.46‰ to +0.14‰

We have not explored the influence of changes in the lifetime of SSA, atmospheric acidity, sea-ice extent so.. The size and sign of the influence that Cl_{MBL} has on $\delta^{13}CH_4$ remains uncertain but we simply cannot ignore it in future interpretations of the glacial-interglacial $\delta^{13}CH_4$ signal

'The role of atomic chlorine in glacial-interglacial changes in the carbon-13 content of atmospheric methane' [Levine et al., 2011b; GRL]

Summary

Sources & Sinks It is plausible the LGM-PI change in [CH_4] was entirely source-driven, and

we can thereby reconcile recent bottom-up model estimates of the changes in CH_4 sources and sinks

(Recent work suggests the rapid rises in [CH_4] at the beginning of D-O events were also source-driven)

Isotopes The size and sign of the influence that Cl_{MBL} has on $\delta^{13}CH_4$ remains uncertain

but we cannot simply ignore it in future interpretations of the glacial-interglacial $\delta^{13}CH_4$ signal

(Of particular interest, could be glacial-interglacial changes in sea-ice extent, and hence SSA production)

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Centre for Atmospheric Science, University of Cambridge (Cambridge p-TOMCAT model)

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