

Supplemental Materials

Table 1. Mass, critical temperature, critical volume and relative diffusion coefficients used for hydrocarbons. D/D_{CO_2} 2 indicates the values used in Worton et al. (2012). Significant length of the scenarios are also provided (in years before drill date).

Species	C ₂ H ₆	C ₃ H ₈	n-C ₄ H ₁₀	i-C ₄ H ₁₀	n-C ₅ H ₁₂	i-C ₅ H ₁₂
Mass	30.07	44.10	58.12	58.12	72.15	72.15
T_{crit}	305.32	369.83	425.12	407.8	469.7	460.4
V_{crit}	145.5	200.	255.	259.	311.	306.
D/D_{CO_2}	0.905	0.702	0.5835	0.5826	0.5051	0.5106
D/D_{CO_2} 2	0.910	0.723	0.615	0.615	0.544	0.544
Length EU	65.7	69.4	72.5	72.5	75.1	74.9
Length US	65.2	68.1	70.6	70.6	72.8	72.6
Length 2009	56.7	60.0	62.7	62.7	-	-

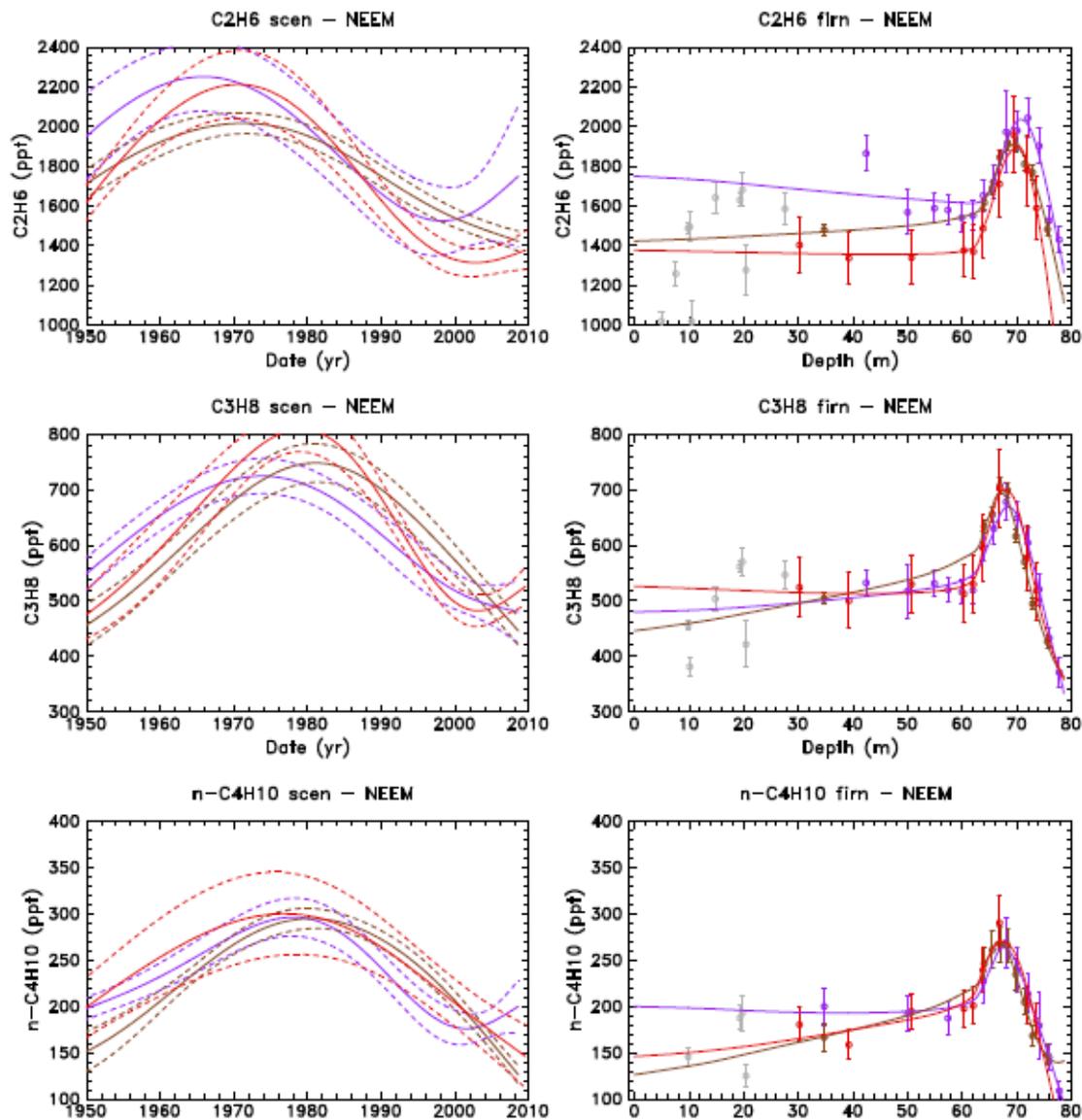


Figure S-1a.

Single site constrained firn profile (right) and atmospheric trend reconstruction (left) for C₂H₆, C₃H₈ and n-C₄H₁₀. Staggered lines represent the 2- σ confidence margins of the model calculations. Colors - NEEM-EU in purple, NEEM-US in brown and NEEM-09 in red. Data points not used for scenario reconstruction (affected by seasonality) are shown in grey. Error bars indicate the 2- σ uncertainty in the data.

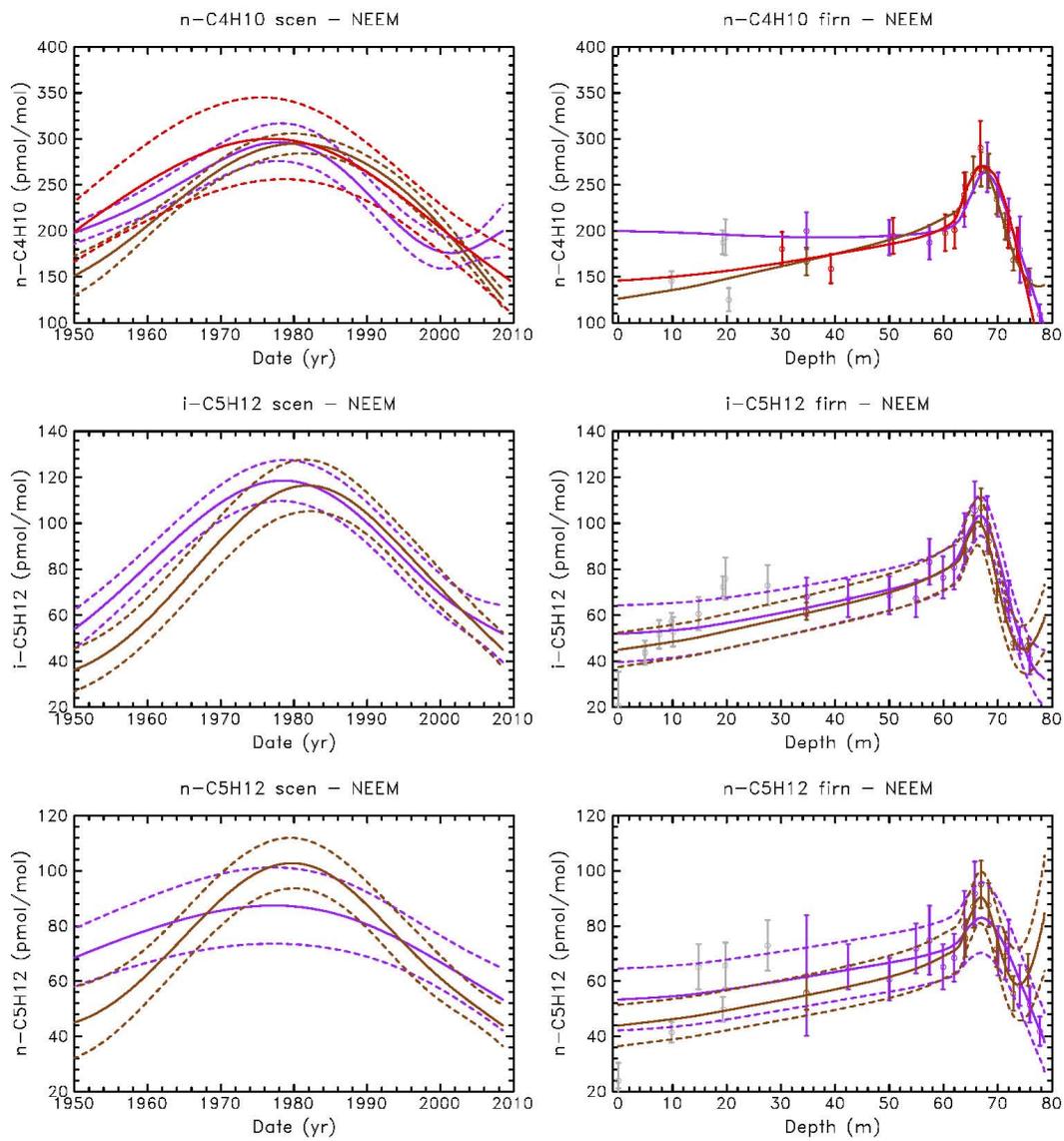


Figure S-1b.
Same as Fig. S-1a for i-C₄H₁₀, n-C₅H₁₂ and n-C₅H₁₂.

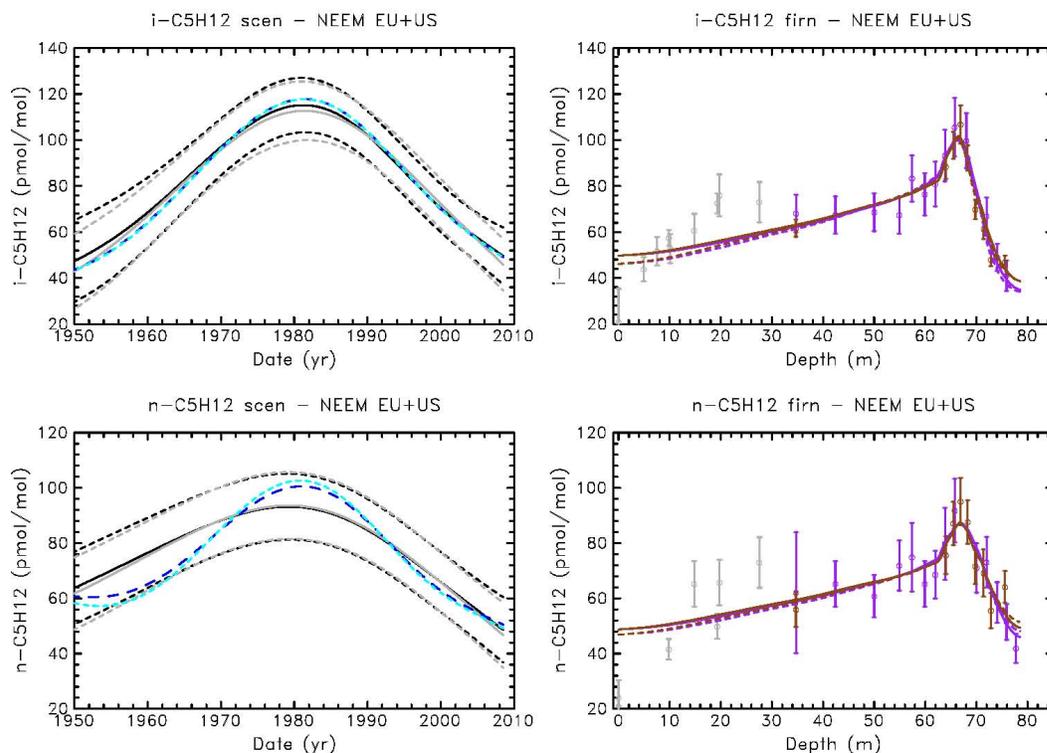


Figure S-2.

Multi-hole constrained atmospheric trend reconstruction for $n\text{-C}_5\text{H}_{12}$ and $i\text{-C}_5\text{H}_{12}$ with INSTAAR model Green's functions. Equal weight for all sites: black lines on left panel and continuous lines on right panel. Weighting using single site optimal RMSD: grey lines on left panel and dashed lines on right panel. Colors for firn results: NEEM-EU in purple, NEEM-US in brown. Scenarios calculated with LGGE-GIPSA model Green's functions are shown in blue for comparison - dark blue: equal weight for all sites, clear blue: weighting using single site optimal RMSD.

Note about Figure S-2:

Results obtained with the INSTAAR model Green's functions for $n\text{-C}_5\text{H}_{12}$ and $i\text{-C}_5\text{H}_{12}$ are consistent within uncertainty limits. They are nearly identical for $i\text{-C}_5\text{H}_{12}$ whereas for $n\text{-C}_5\text{H}_{12}$ the INSTAAR model Green's functions produce a smoother scenario than the LGGE-GIPSA model Green's function. This difference can be better understood by comparing the right panels of Figure S-2 and Figure 5b of the main paper. The LGGE-GIPSA model Green's functions produce a slightly narrower $n\text{-C}_5\text{H}_{12}$ peak in the firn with a slightly better match of the firn data at 60-64 m depth and a more complex shape of the deep firn profile (below 70 m depth). These differences are not significant in comparison with the uncertainties on the data (which are comparable to the scenario uncertainty envelope).

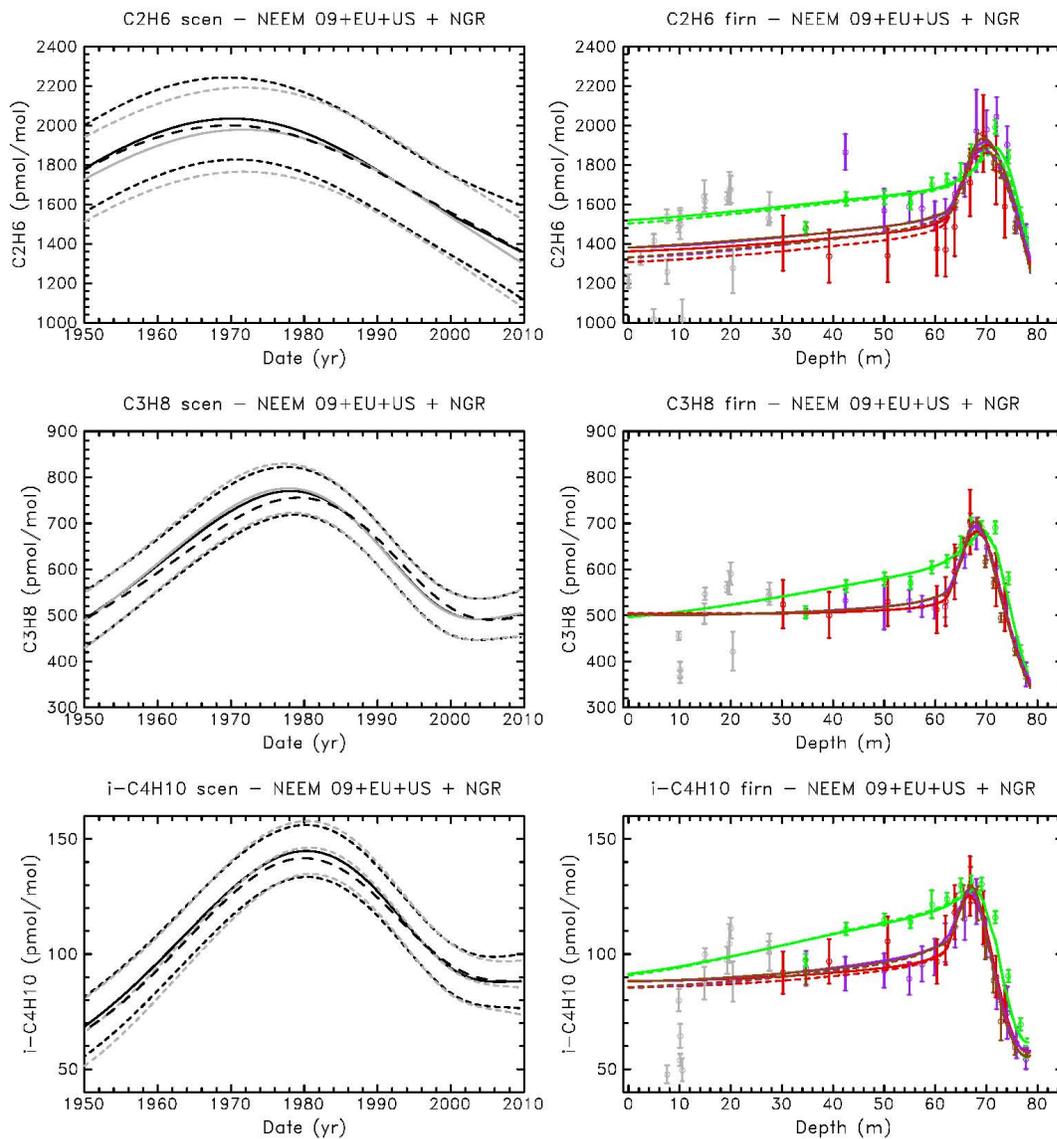


Figure S-3a.

Multi-site constrained atmospheric trend reconstruction with North GRIP and NEEM for C₂H₆, C₃H₈ and *i*-C₄H₁₀. Equal weight for all boreholes: black lines on left panel and continuous lines on right panel. NEEM only scenario with equal weight for all boreholes: black long dashed lines in left panels. NEEM + North GRIP constrained simulations with weighting using single borehole optimal RMSD: grey lines in left panels and dashed lines in right panels. Colors for firm results: North GRIP in green, NEEM-EU in purple, NEEM-US in brown, NEEM 2009 in red.

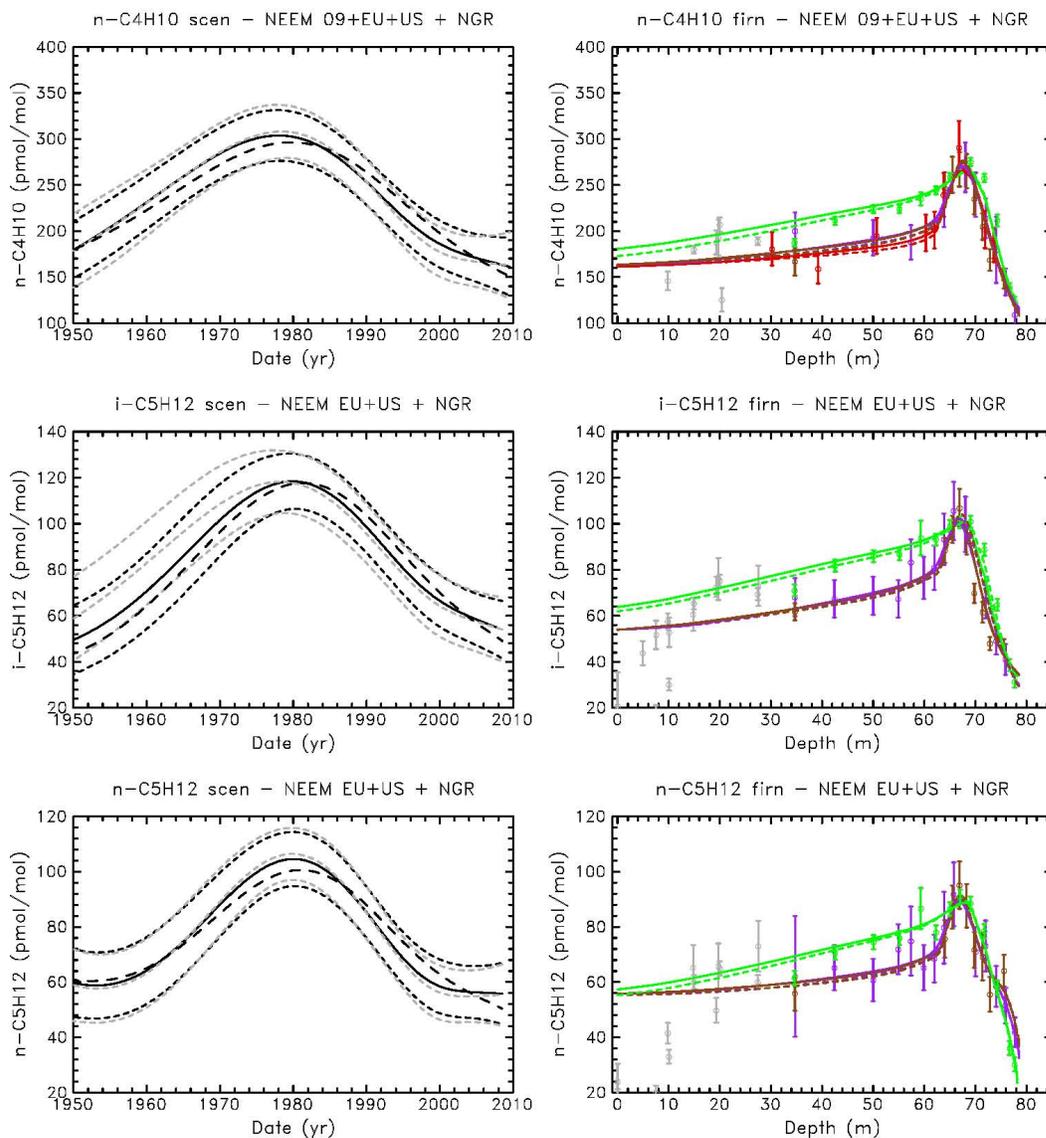


Figure S-3b

Same as Fig. S-3a for n-C₄H₁₀, i-C₅H₁₂ and n-C₅H₁₂

Note about Figure S-3:

The North GRIP and NEEM data are not fully consistent in terms of calibration scale and methodology for uncertainty evaluation. However North GRIP brings a new constraint to the model with respect to NEEM because it was drilled 7-8 years earlier (2001). Directly comparing the North GRIP and NEEM firn data, the most striking difference lies in the slope and concentration levels in the upper firn (30 m to 60 m range), which reflects this difference in drill dates. The NEEM only and NEEM + North GRIP constrained scenarios are consistent (within

uncertainty limits of one another) and no strong increase in the widths of the scenario uncertainty envelopes is observed, indicating a good consistency of the NEEM and North GRIP data.

