

Figure S1: Methane mixing ratios at the RV Helmer Hanssen vs apparent wind angle relative to the bow (β), using $\beta = \arccos(W \cos \alpha + V / \sqrt{W^2 + V^2 + 2WV \cos \alpha})$ where α =true pointing angle, W =true wind velocity, and V =ship velocity.

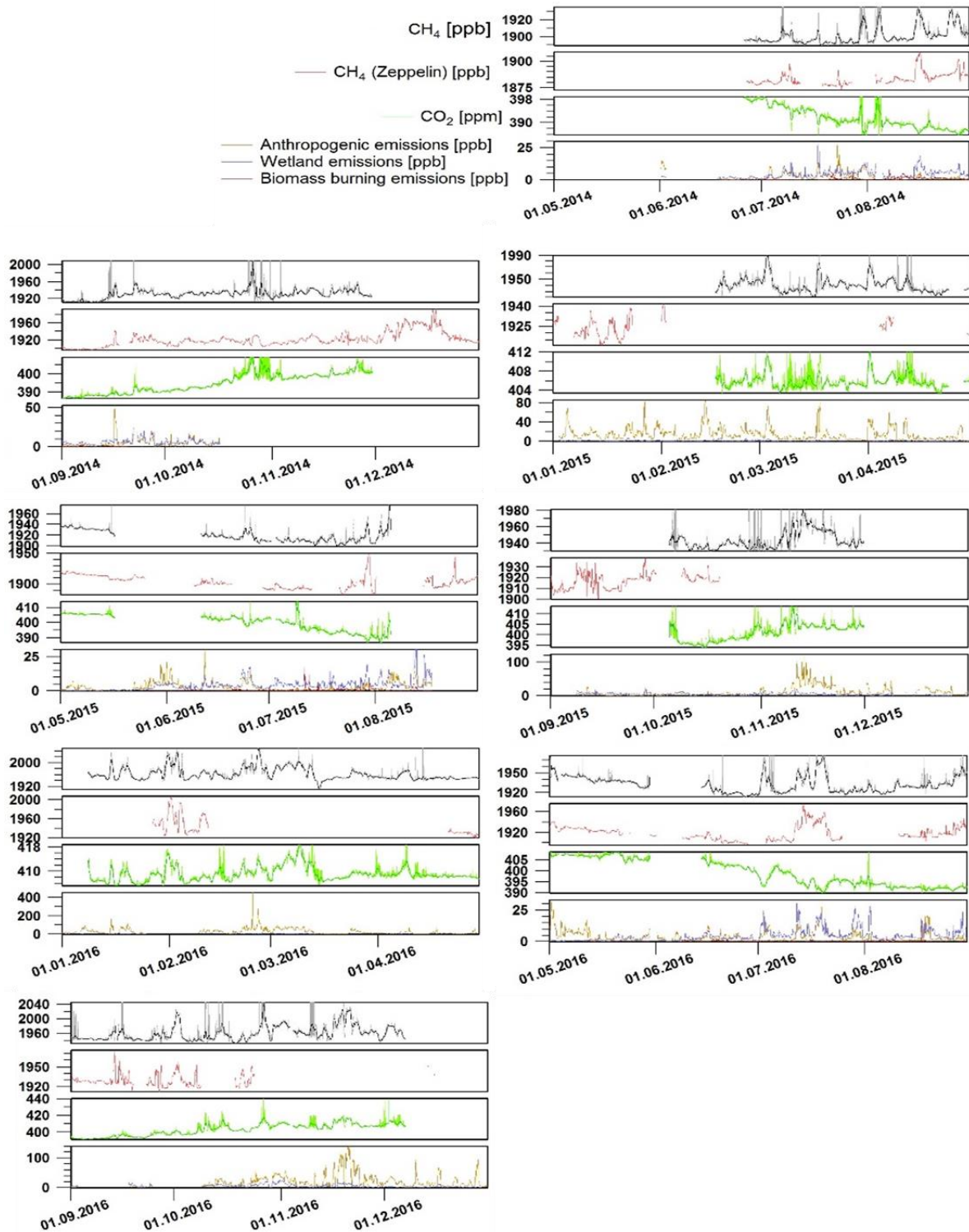
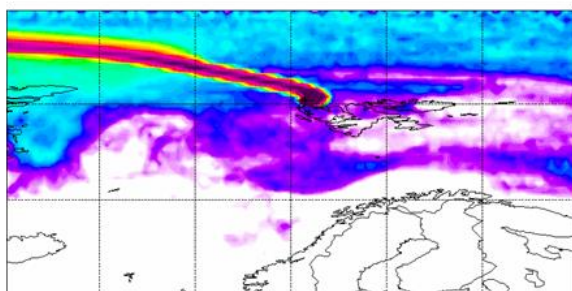
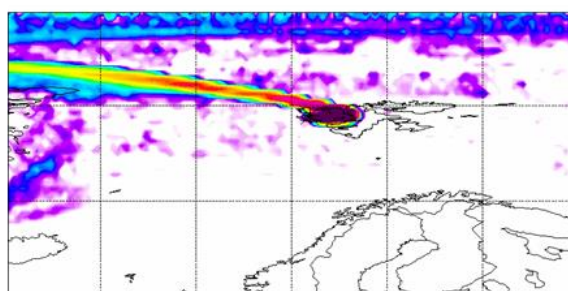


Figure S2: Long term, continuous time series and model data presented in this study. From top to bottom: Methane (CH₄) concentrations measured at the RV Helmer Hanssen (1 minute, light grey) and box car –smoothed (dark grey) for easier visualisation of major excursions from baseline concentrations, CH₄ at the Zeppelin Observatory (hourly), for ship positions within 75-82° N, 5-35° E (pink), Carbon dioxide (CO₂) at the RV Helmer Hanssen (1 min, light green) and smoothed (dark green), and terrestrial emissions from anthropogenic activity (gold), wetlands (blue) and biomass burning (brown) according to emission inventories and FLEXPART (see text for details, hourly or less). Note, the Y-axis range changes on each panel.

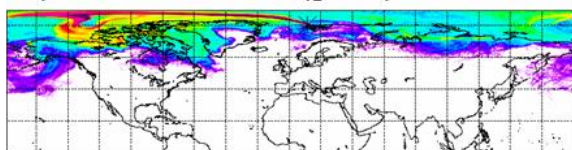
A) RV Helmer Hanssen (regional)



B) Zeppelin Observatory (regional)



C) RV Helmer Hanssen (global)



D) Zeppelin Observatory (global)

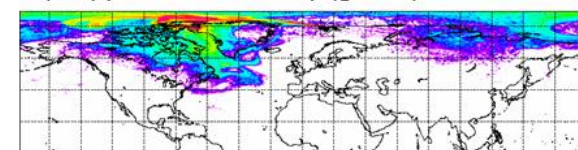


Figure S3: FLEXPART footprint sensitivities on 25.08.2014 for the RV Helmer Hanssen and Zeppelin Observatory. A) the RV Helmer Hanssen at 5: 32 AM at a regional scale B) Zeppelin Observatory at 6:00 AM, C) Same as A), global scale, D) Same as B), global scale.

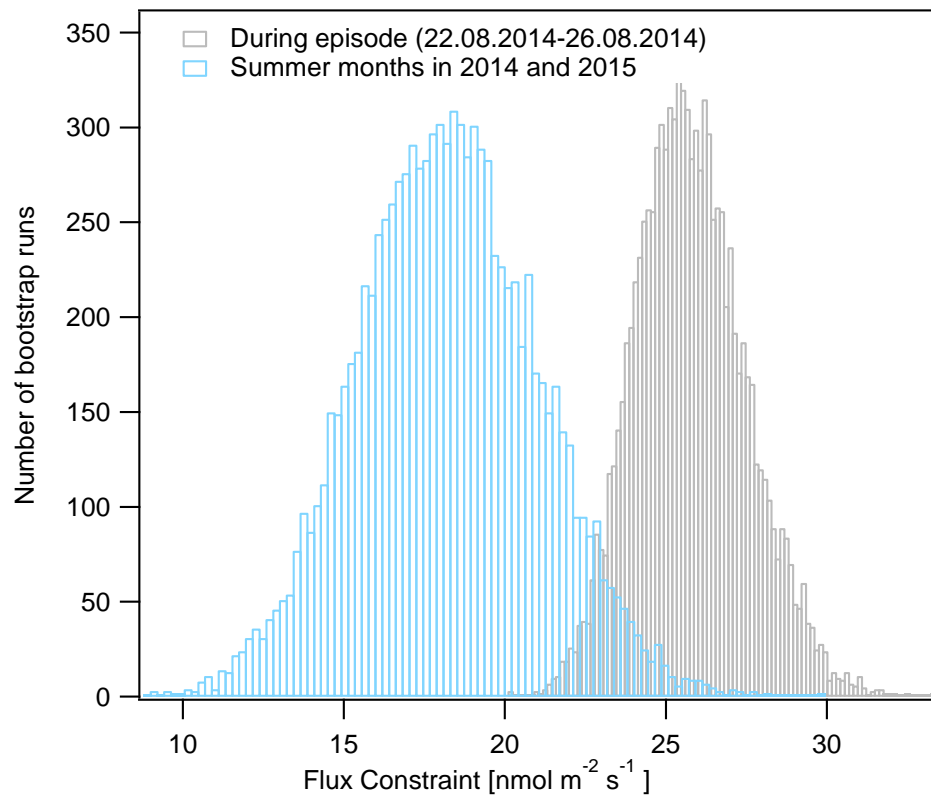


Figure S4: Distribution of results (100 bins) obtained from bootstrapping ($n=10000$) the methane mixing ratios measured at the RV Helmer Hanssen and mean FLEXPART footprint sensitivity to the area $80.39\text{-}81.11^{\circ}\text{N}, 13.83\text{-}19^{\circ}\text{E}$ (north of Svalbard) and calculating a flux constraint for methane based on the upwind-downwind methodology of (Pisso et al., 2016). Data in blue show results from performing the analysis for the time periods 19.06.2014-31.08.2014 and 01.06.2015-31.08.2015 combined (i.e. summer months). Data in grey show the distribution of the calculated flux constraints for 22.08.2014-28.08.2014.

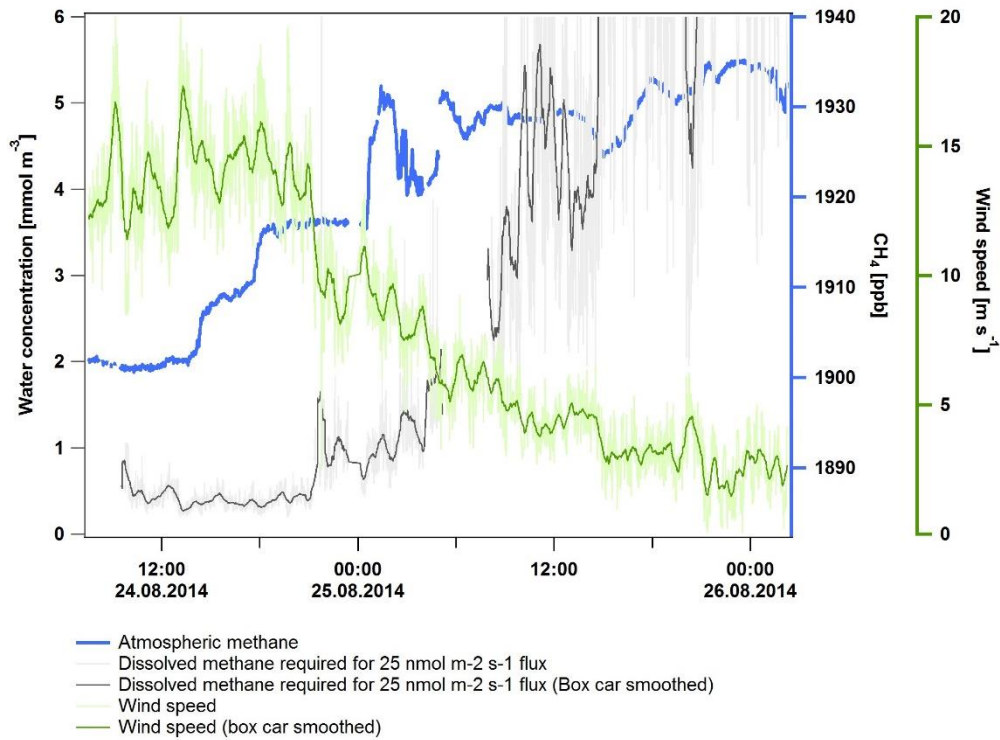


Figure S5: Atmospheric methane (CH₄, blue), wind speed (green) and estimated dissolved CH₄ required to produce the observed changes in atmospheric CH₄ during an episode of high CH₄ concentrations observed north of Svalbard.

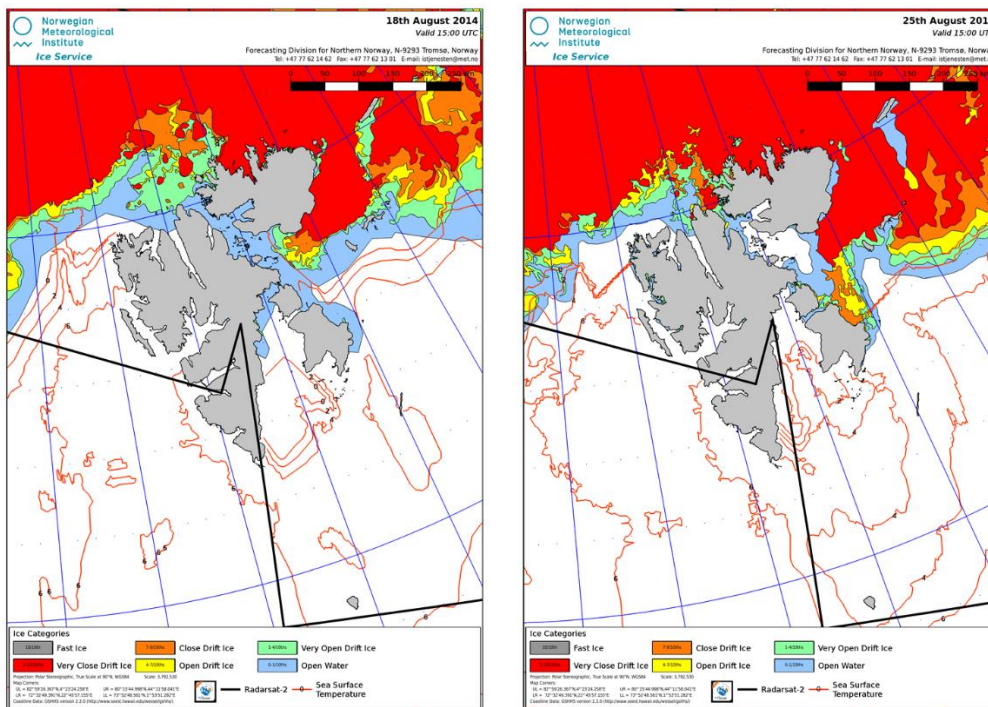


Figure S6: Ice cover around Svalbard on 18th and 25th August 2014, from the Norwegian Ice Service of the Norwegian Meteorological Institute (Klimadata: <https://klimaservicesenter.no>).

