

Supplemental Information

Section S1. Model Comparison of Independent Sites

Here we show the comparison between the model and surface observations at the independent observation sites that have not been used in the assimilation. The model captures the SCA amplitude well when compared with surface observations (Fig. S1). The Siberia sites (DEM, BRZ and KRS) have a very short observation record of 6 years and the SCA is quite variable within this time period, leading to large error bars.

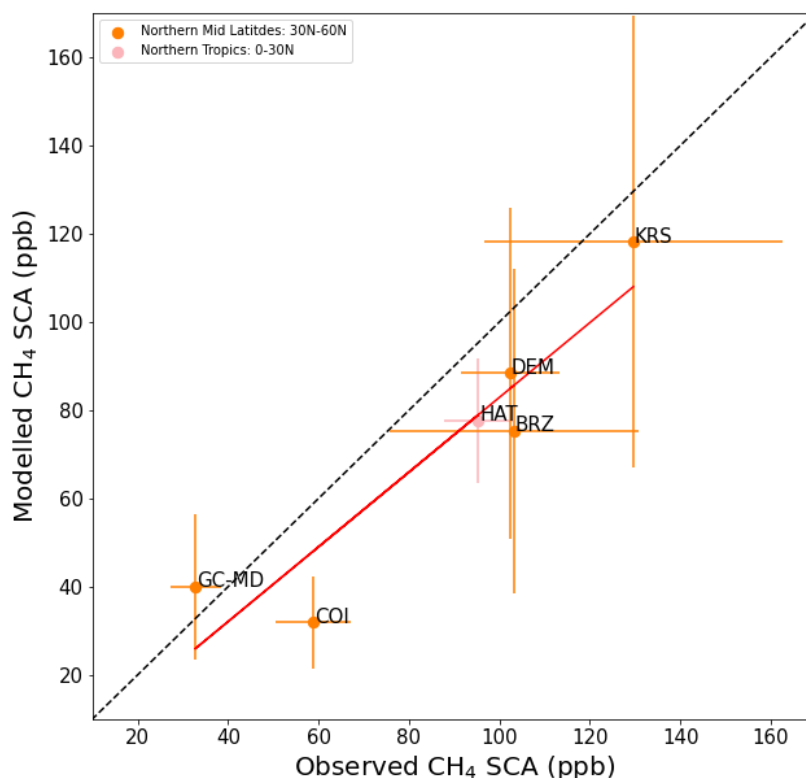


Figure S1: The comparisons between simulated and observed CH₄ at the independent surface observation sites. The solid line represents the unit lie and the red line represents the least squares regression line. The error bars denote $\pm 1\sigma$.

Table S1. List of sites used in the independent analysis

Site name	Latitude (°N)	Longitude (°E)	Site code
Cape Ochi-ishi, Japan	43.16	145.45	COI
Hateruma, Japan	24.06	123.80	HAT
Demyanskoe, Russia	59.79	70.87	DEM
Karasevov, Russia	58.25	82.42	KRS
Berezorechka, Russia	56.15	84.33	BRZ
Mace Head, Republic of Ireland	53.33	-9.90	GC-MD

Section S2. SCA Time Series

This section contains the time series of the modelled and observed SCA and the trend at the NOAA observation sites (Fig. S2)

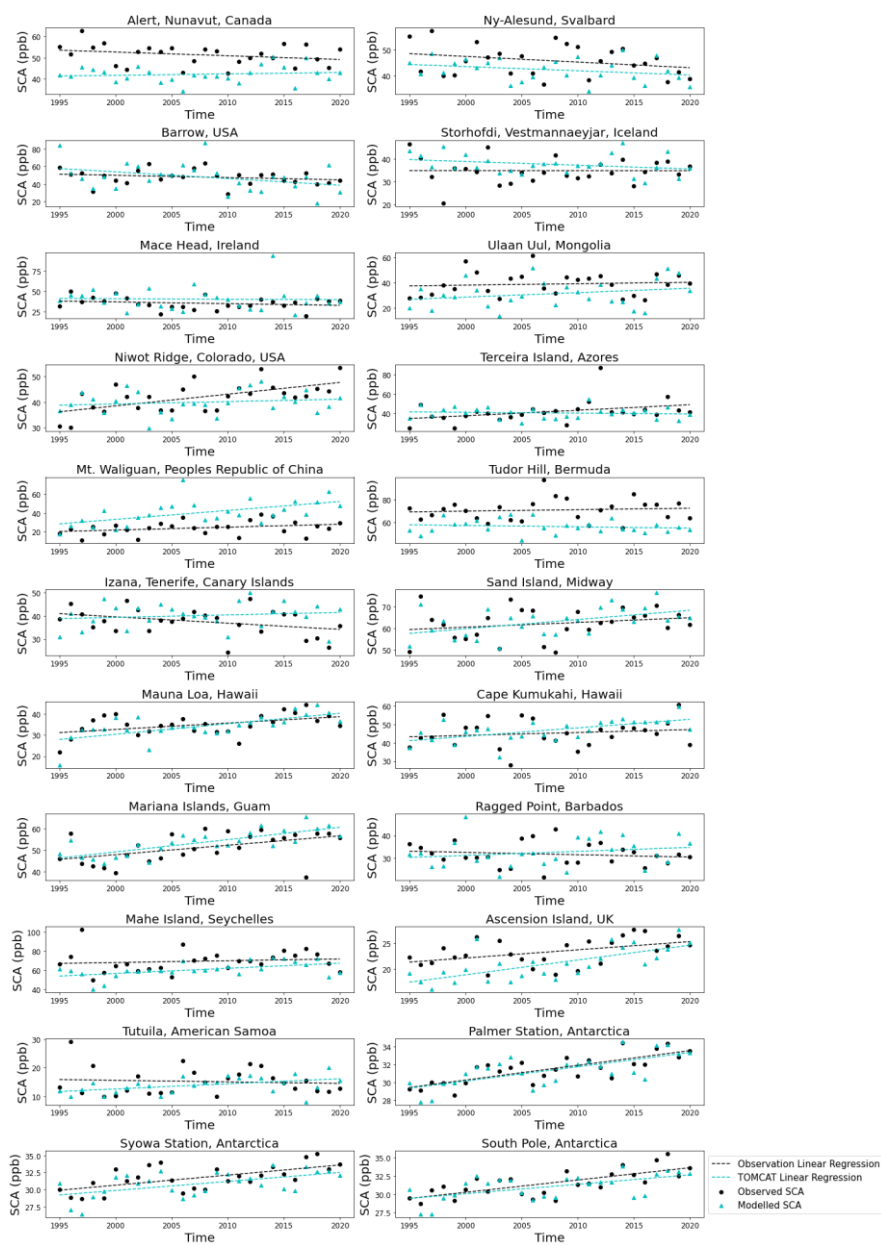


Figure S2: The time series comparison of modelled and observed SCA and the trend at the NOAA observation sites. The sites are ordered north to south.

Section S3. Regional Contribution in the NHL

This section contains the regional contributions under different scenarios. Figure S3 shows the normalised contributions of the regional tagged tracers to show the impact of the region size on the results. Figure S4 shows the contributions of the tagged tracers without BRW in the NHL regions. Figure S5 shows the contributions of the tagged tracers with MHD in the NHL which is classed as 52N-60N in this case.

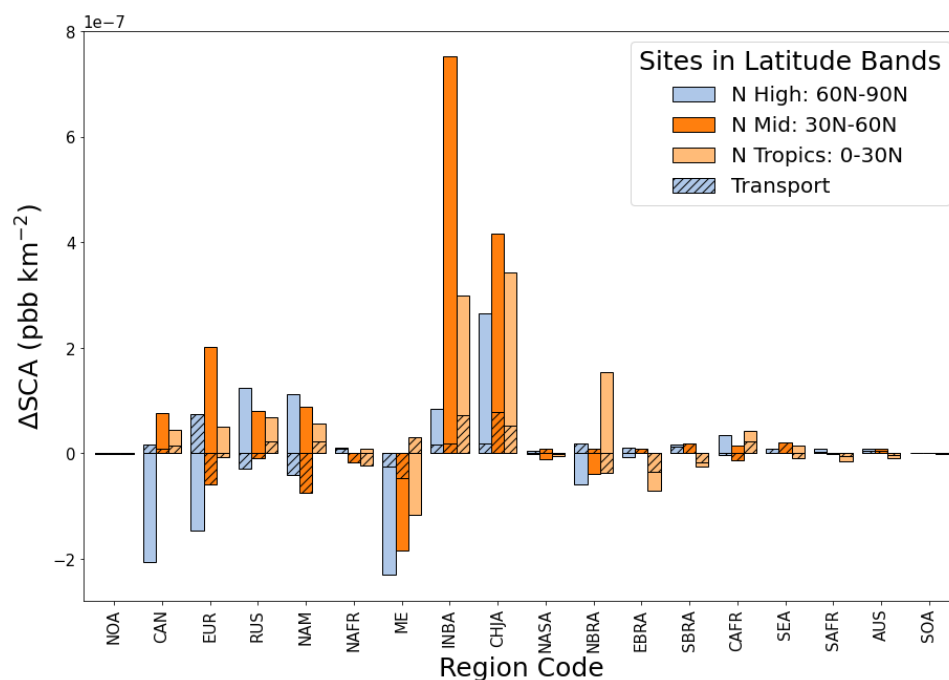


Figure S3: The normalised contribution of the regional tagged tracers to CH_4 ΔSCA (ppb km^{-2}) as a mean across all sites in the latitude band. The hatched bars show the contribution from transport (TOM_transport) and the solid colour represents the contribution from emissions (TOM_regional).

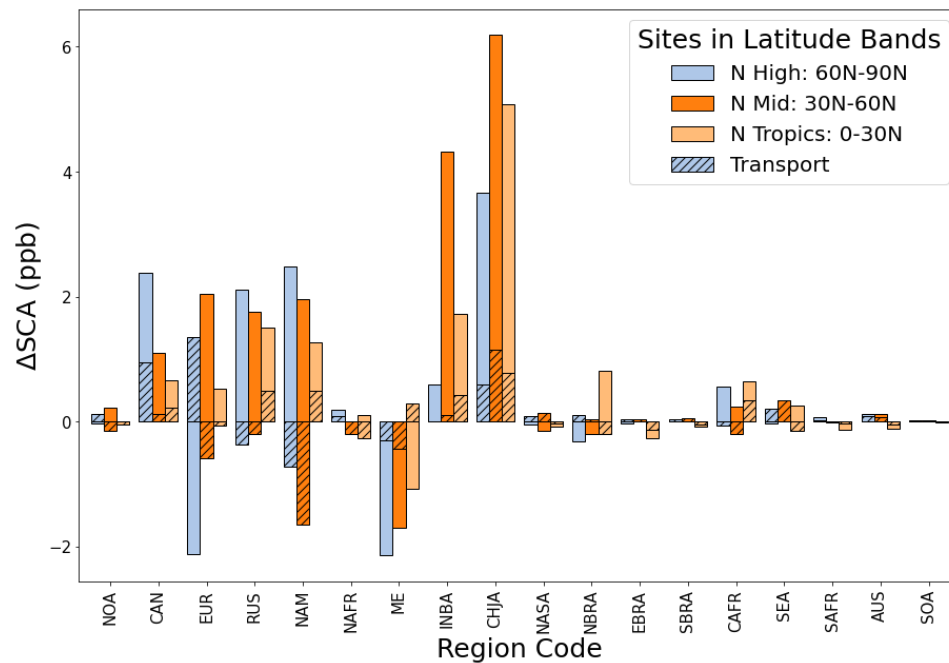


Figure S4: The contribution of the regional tagged tracers to CH₄ ΔSCA (ppb) as a mean across all sites in the latitude band. The hatched bars show the contribution from transport (TOM_transport) and the solid colour represents the contribution from emissions (TOM_regional). The site at BRW has been removed in the NHL.

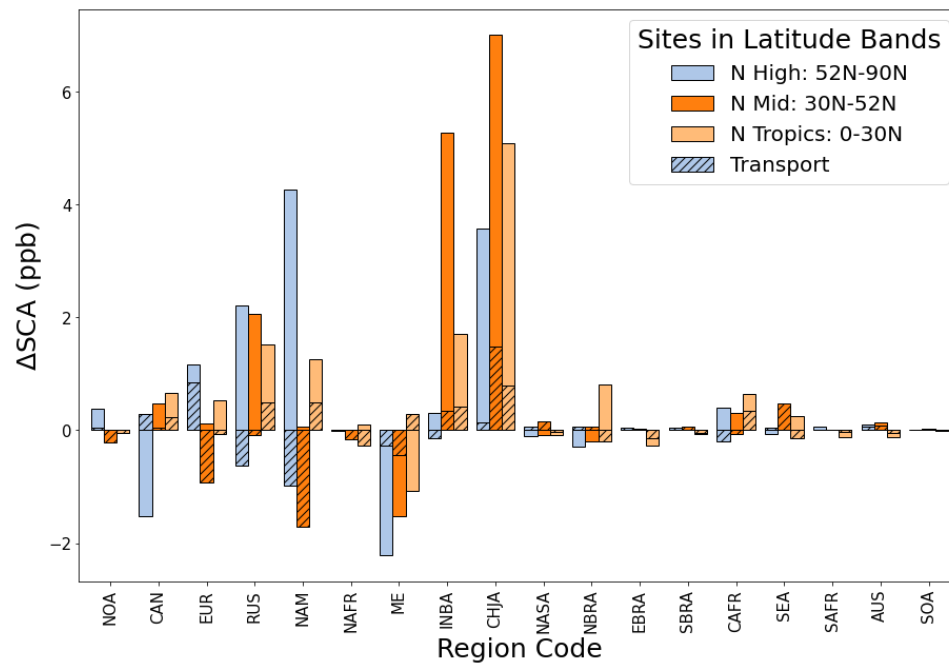


Figure S5: The contribution of the regional tagged tracers to CH₄ ΔSCA (ppb) as a mean across all sites in the latitude band. The hatched bars show the contribution from transport (TOM_transport) and the solid colour represents the contribution from emissions (TOM_regional). MHD has been added to the NHL region (52N-90N).

Section S3. Mean Emission Maps & Trends

This section contains the seasonal mean emissions for the regions which contribute the most to the Δ SCA in the NHL. It also contains the seasonal trends of emissions in each of these regions.

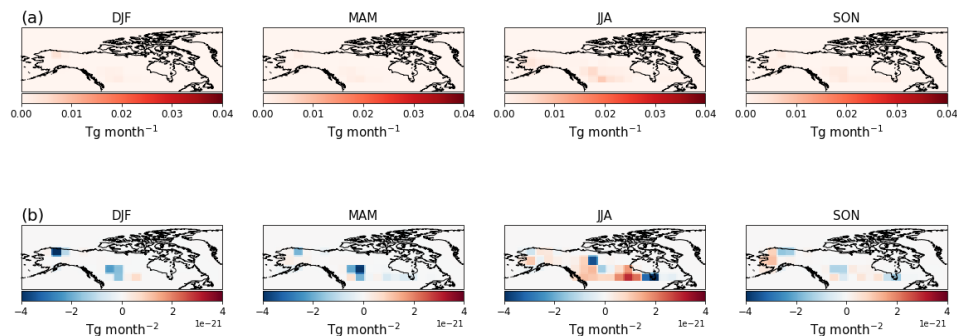


Figure S6: (a) Seasonal mean emissions ($Tg\ month^{-1}$) and (b) seasonal trend in emissions ($Tg\ month^{-2}$) over Canada between 1995-2020.

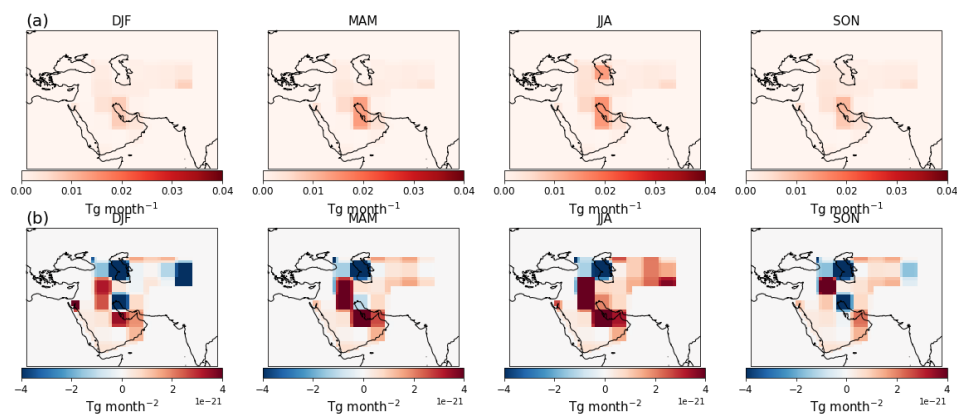


Figure S7: (a) Seasonal mean emissions ($Tg\ month^{-1}$) and (b) seasonal trend in emissions ($Tg\ month^{-2}$) over Middle East between 1995-2020.

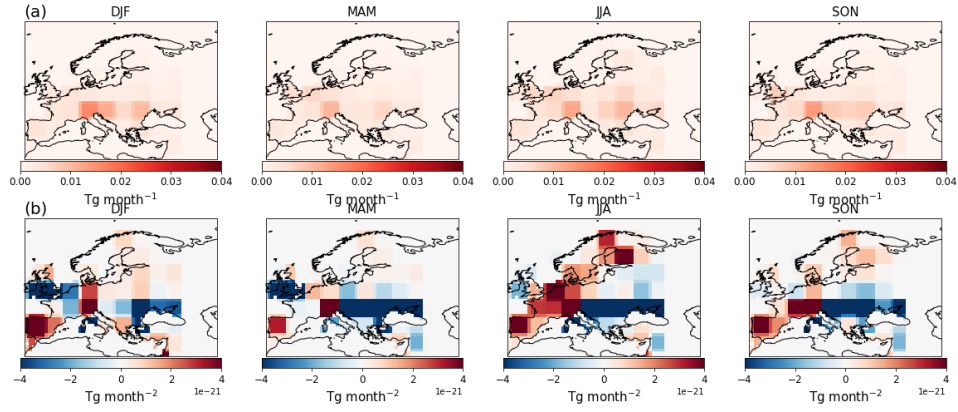


Figure S8: (a) Seasonal mean emissions (Tg month^{-1}) and (b) seasonal trend in emissions (Tg month^{-2}) over Europe between 1995-2020.

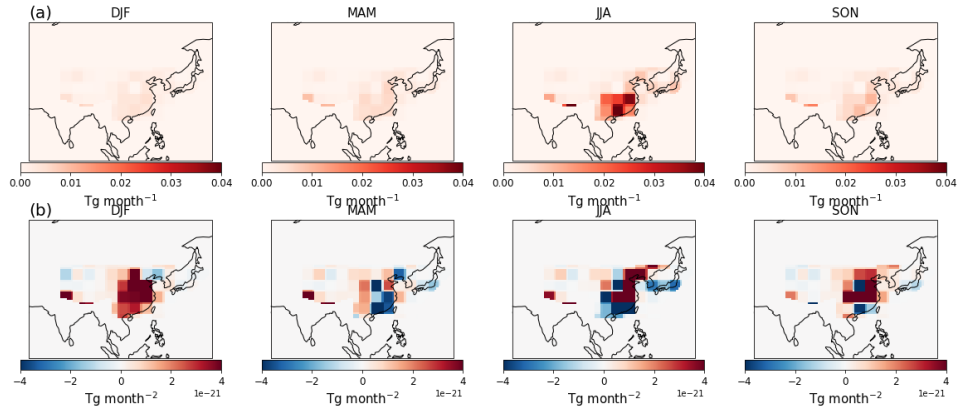


Figure S9: (a) Seasonal mean emissions (Tg month^{-1}) and (b) seasonal trend in emissions (Tg month^{-2}) over China & Japan between 1995-2020.

Section S4. GFED Fire Emissions

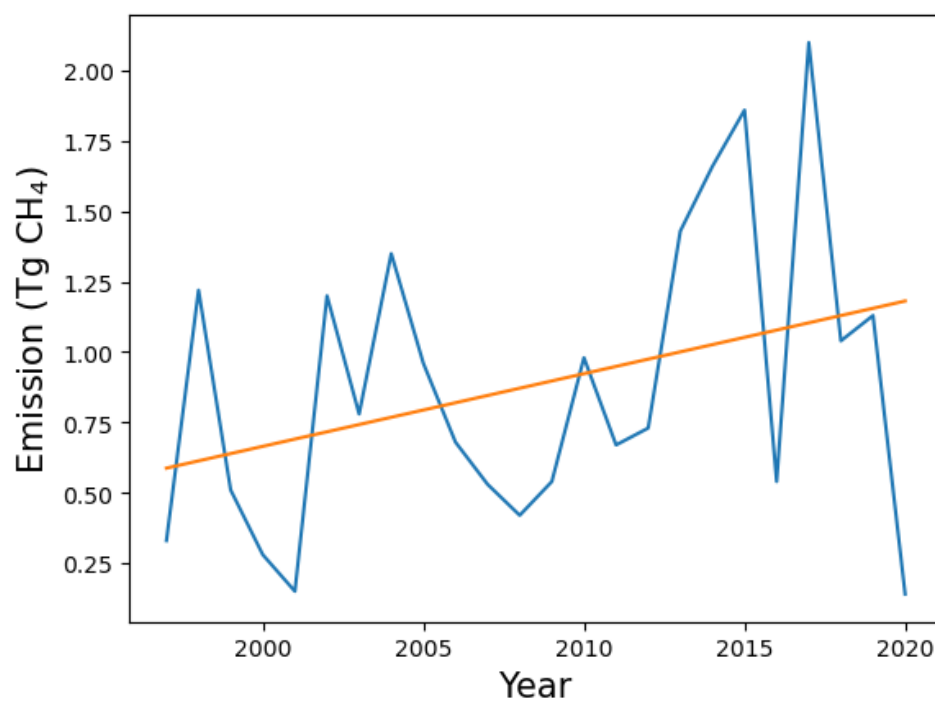


Figure S10: Emissions of CH₄ (Tg CH₄) from fires over Canada between 1997-2020 (blue) and its trend (orange).

Section S5. Regional Concentration Contributions to Northern High Latitudes

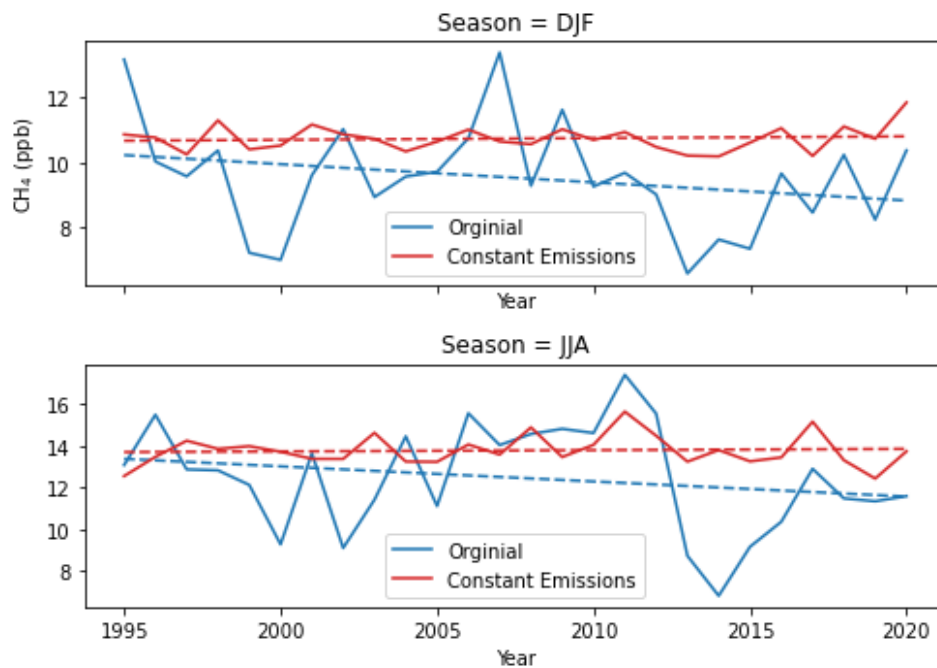


Figure S11: Mean CH_4 (ppb) contribution across the NHL at the surface (60N-90N) for the TOM_regional simulation (blue) and the TOM_transport simulation (red) from Canada.

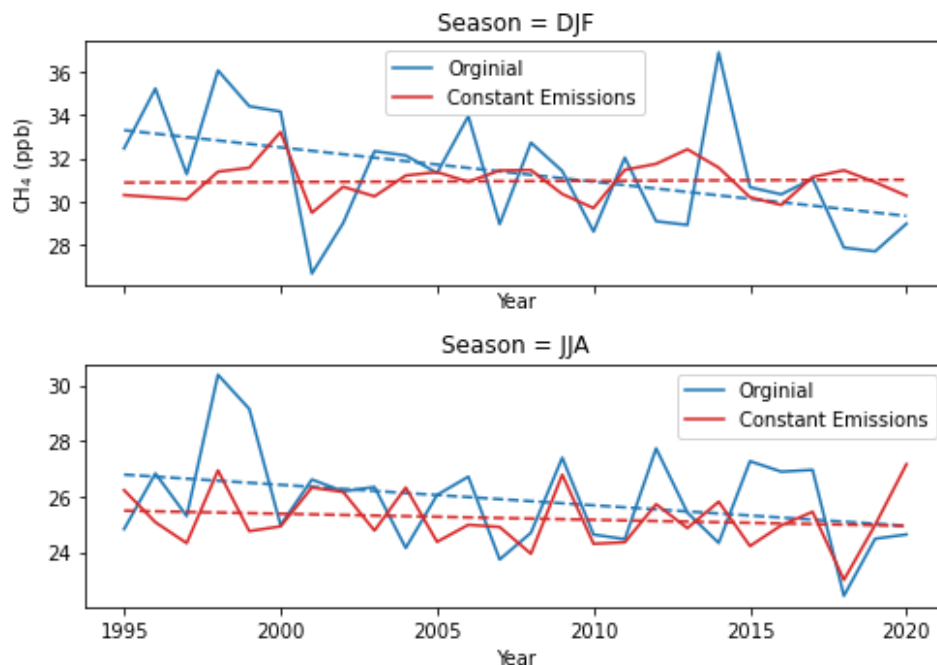


Figure S12: Mean CH_4 (ppb) contribution across the NHL at the surface (60N-90N) for the TOM_regional simulation (blue) and the TOM_transport simulation (red) from Europe.

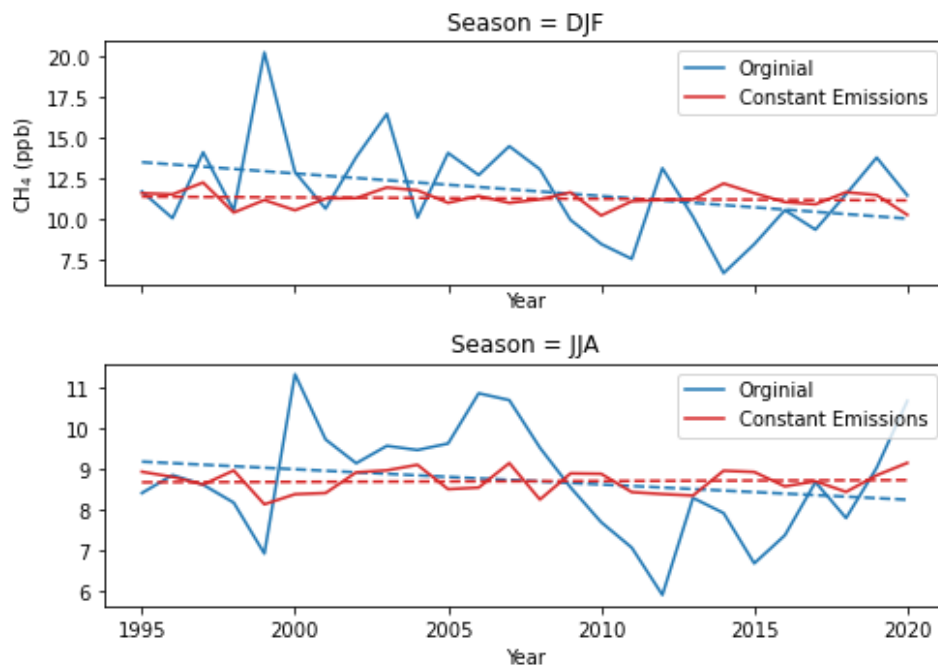


Figure S13: Mean CH_4 (ppb) contribution across the NHL at the surface (60N-90N) for the TOM_regional simulation (blue) and the TOM_transport simulation (red) from the Middle East.

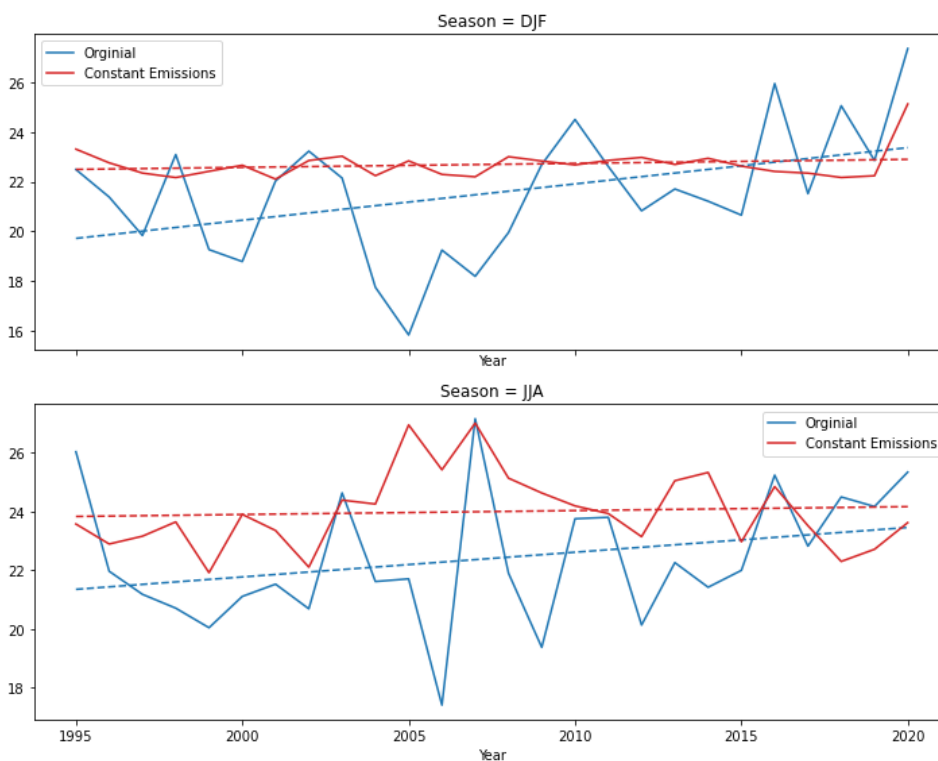


Figure S14: Mean CH_4 (ppb) contribution across the NHL at the surface (60N-90N) for the TOM_regional simulation (blue) and the TOM_transport simulation (red) from China & Japan.