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Importance of seasonally resolved oceanic emissions for bromoform delivery from the tropical Indian Ocean and west Pacific to the stratosphere

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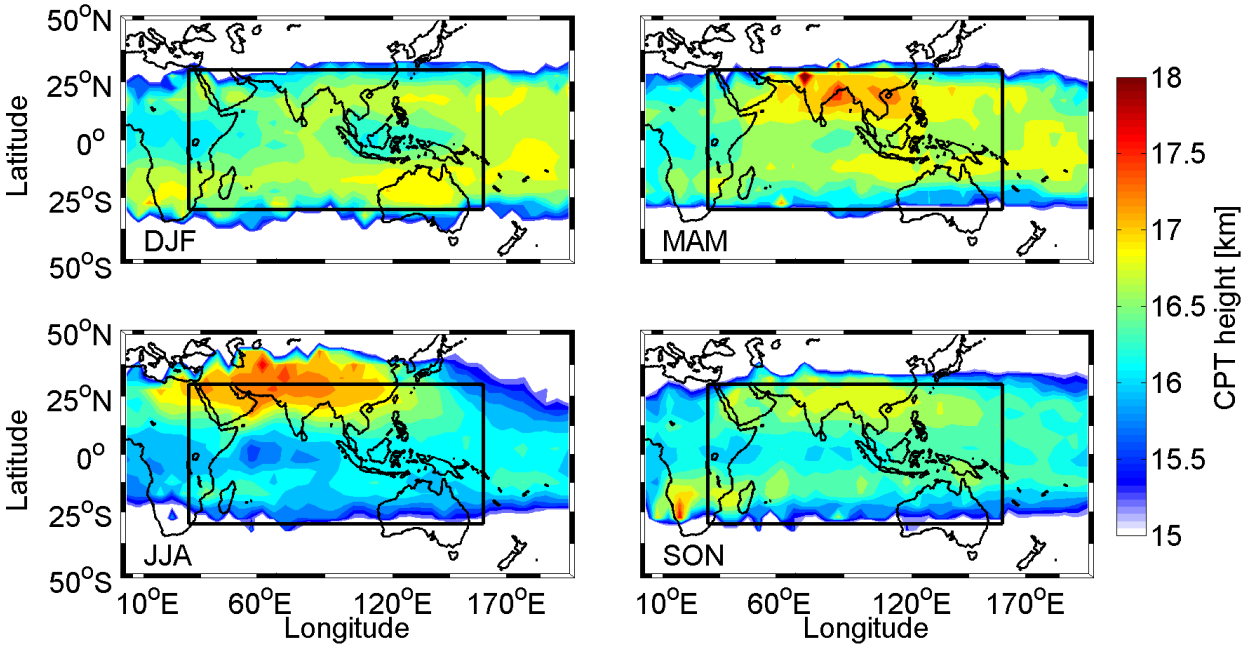


Figure S1: Height of the cold point tropopause (CPT) for different seasons from December 2013 to November 2014 from ERA-Interim data used in FLEXPART.

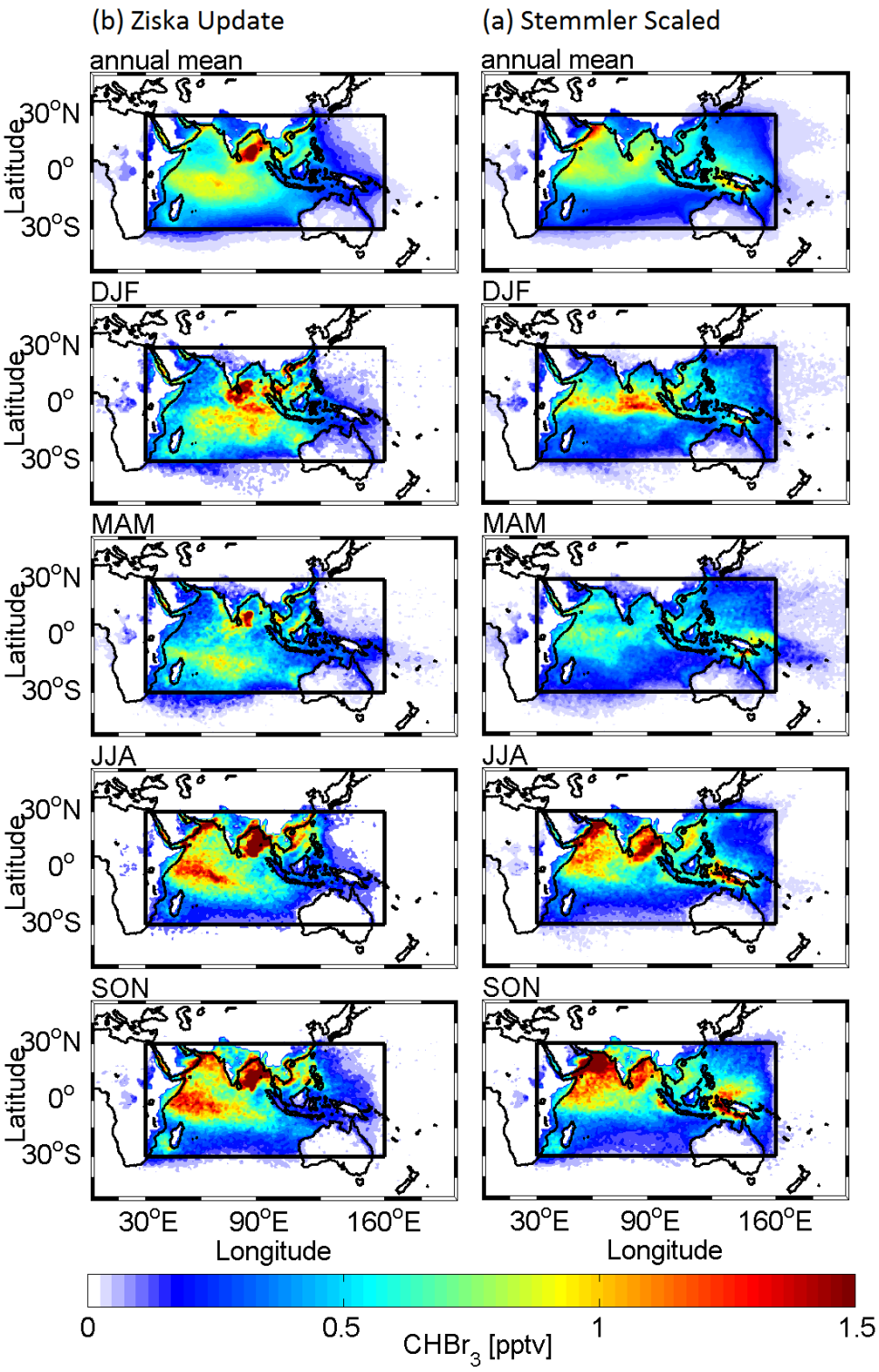
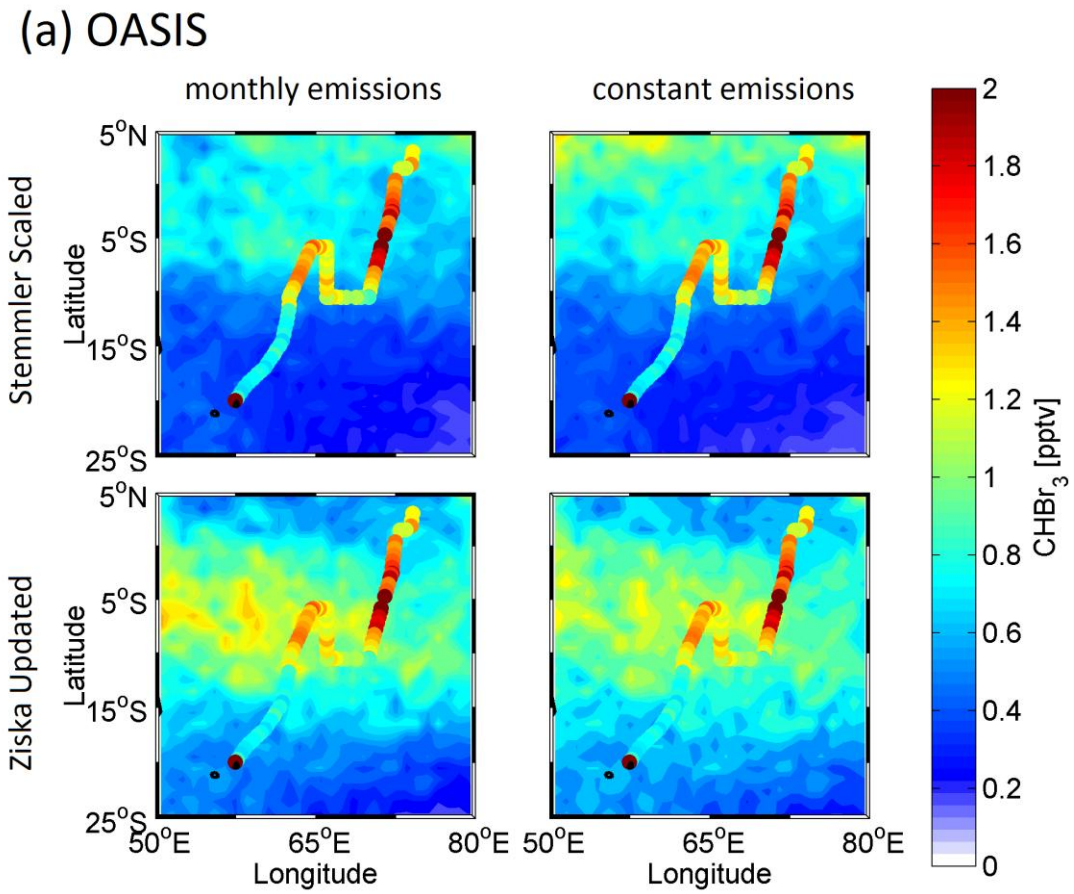


Figure S2: Bromoform volume mixing ratios (VMR) at 100 m height for monthly emissions from (a) Ziska Updated and (b) Stemmler Scaled inventories with monthly emissions. The black box depicts the Indian Ocean/West Pacific release area.

Model-measurement comparison (Fig. S3)

In the Indian Ocean, volume mixing ratios (VMR) using the Ziska Updated inventory are closer to observations than the Stemmler Scaled inventory, because the OASIS cruise data was included directly into the Ziska Updated inventory, and monthly averaged emissions fit better than annually averaged emissions because of the pronounced annual cycle in the Indian Ocean emissions (Fig. S3a). In the tropical west Pacific, the Stemmler Scaled inventory has a smaller deviation from the observations than the VMR modeled using Ziska Updated emissions, because of the low oceanic concentrations during TransBrom in October 2009 included in Ziska Updated inventory (Fig. S3b). In the West Pacific, the difference between monthly and annually averaged emissions in boreal fall is smaller than in the Indian Ocean during boreal summer. In the South China Sea, VMR modeled from both inventories are similar, but far lower than the coastal SHIVA cruise observations, probably because of underestimation of coastal emissions in both inventories (Fig. S3c).



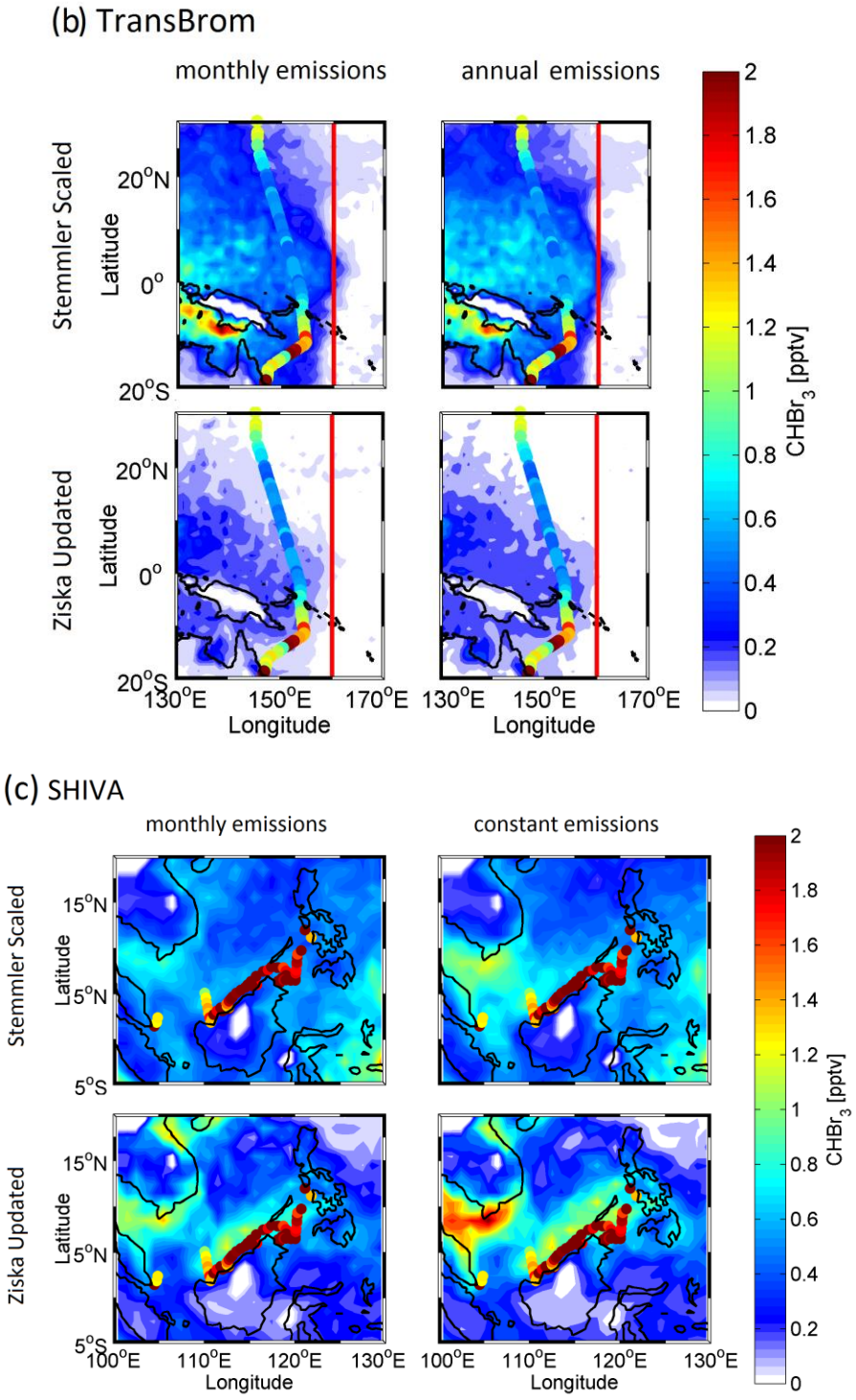


Figure S3: Comparison of modeled bromoform volume mixing ratios (VMR) at 100 m height and ship cruise measurements in the (a) Indian Ocean in July 2014 (OASIS), (b) the west Pacific in October 2009 (TransBrom), and (c) South China and Sulu Sea in November 2011 (SHIVA). The modeled VMR is displayed for the two emission inventories Stemmler Scaled and Ziska Updated and the monthly and annually averaged emission scenarios in the month of the respective cruise but for the year 2014. The red line in (b) displays the edge of the IO/WP release area.

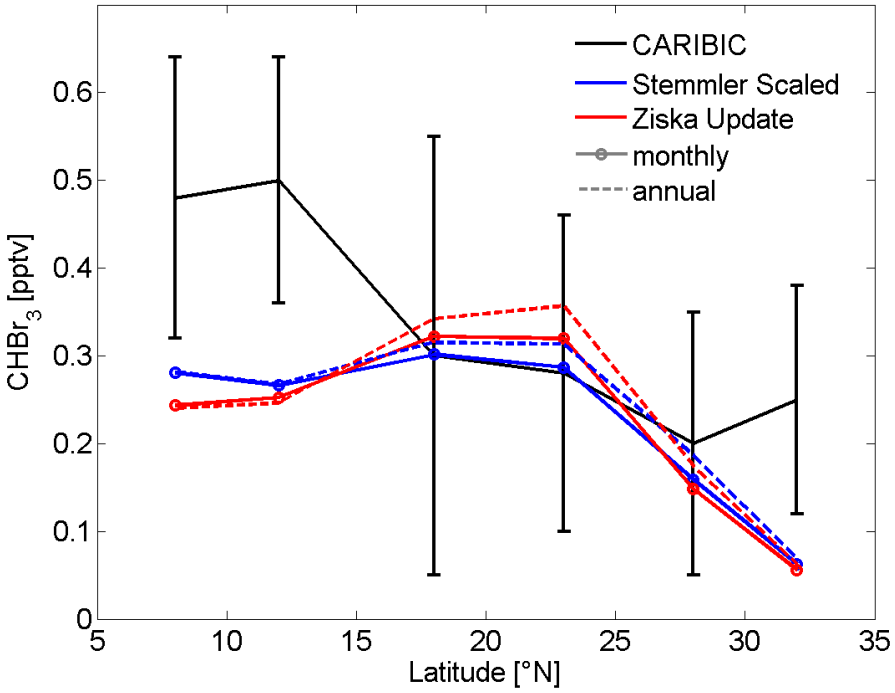


Figure S4: CARIBIC aircraft measurements of bromoform volume mixing ratio (VMR) on Southeast Asian (70°E -100°E) flights for November and December 2012 and February 2013 at about 11 km height averaged in 5° latitude bins taken from Wisher et al. (2014). Stemmler Scaled and Ziska Update $CHBr_3$ VMR at 11 km averaged in 5°x5° boxes around the measurement locations in February, November, and December of 2014.

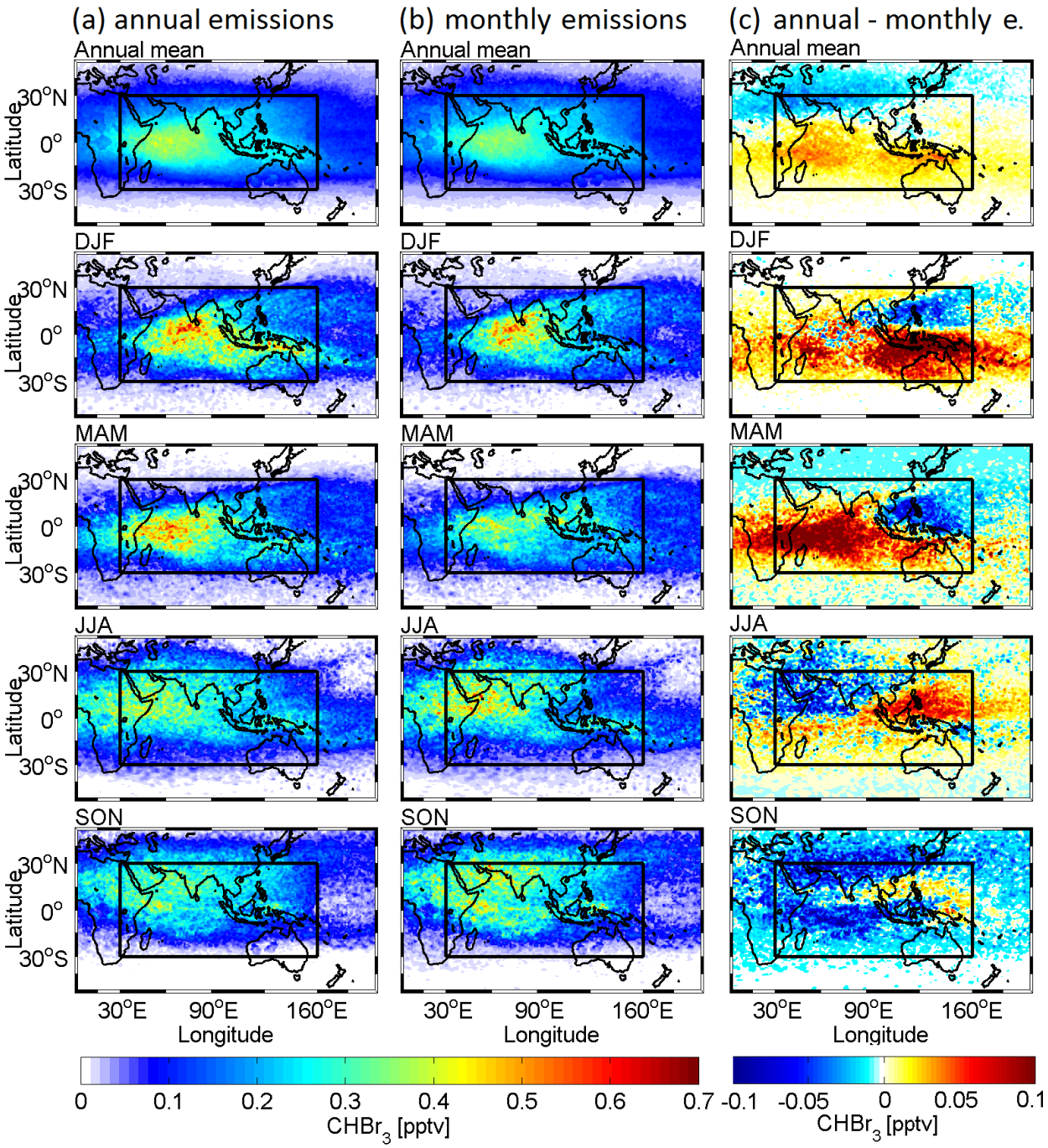


Figure S5: Bromoform volume mixing ratios (VMR) at 17 km for the Stemmler Scaled (a) annually and (b) monthly averaged emissions and (c) the difference between the two scenarios. The black box depicts the Indian Ocean/West Pacific release area.

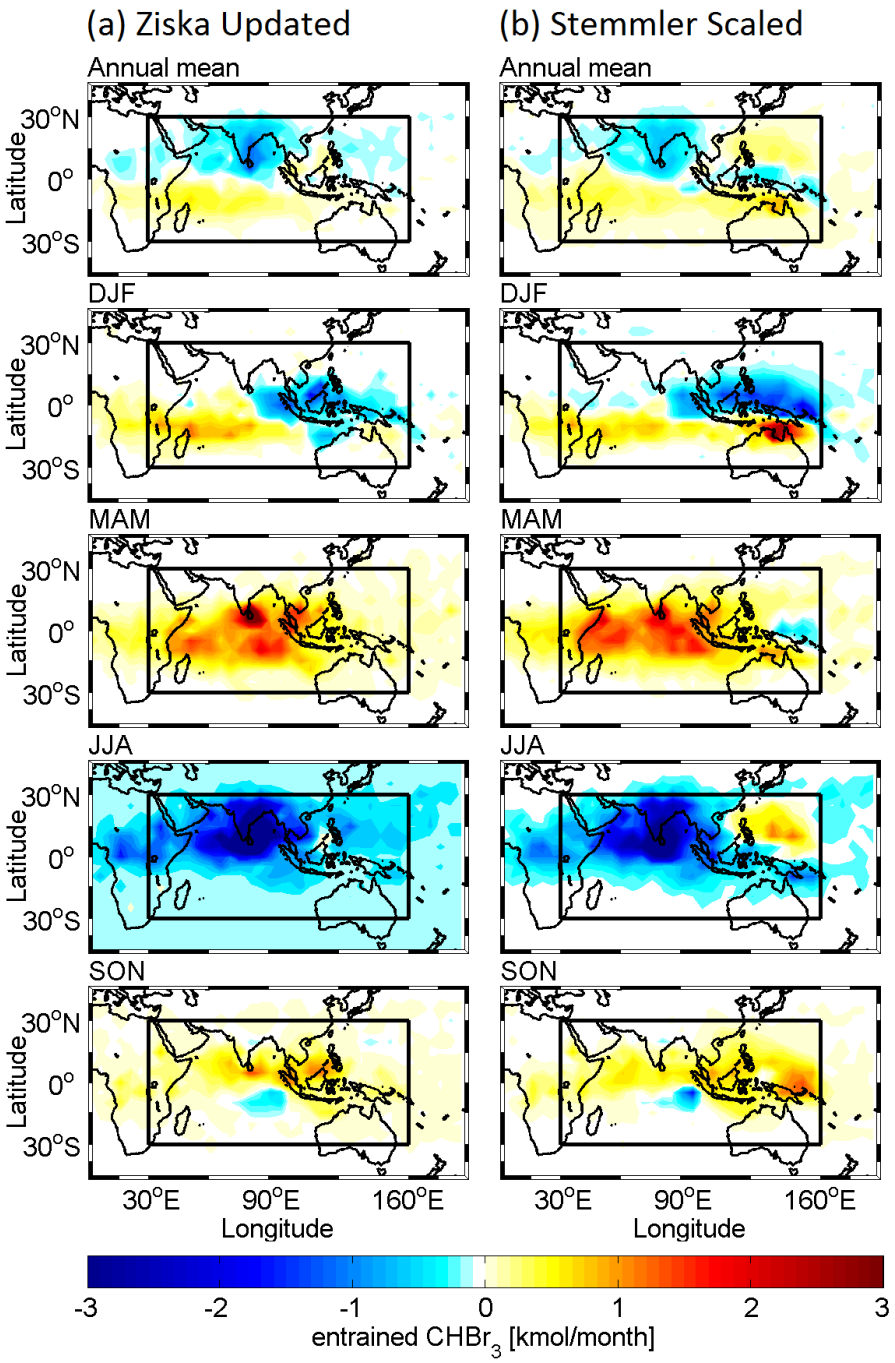


Figure S6: Bromoform entrainment anomalies at the cold point tropopause (CPT) between annually and monthly averaged emissions for (a) Ziska Updated and (b) Stemmler Scaled inventories.

References

Wisher, A., Oram, D. E., Laube, J. C., Mills, G. P., van Velthoven, P., Zahn, A., and Brenninkmeijer, C. A. M.: Very short-lived bromomethanes measured by the CARIBIC observatory over the North Atlantic, Africa and Southeast Asia during 2009–2013, Atmos. Chem. Phys., 14, 3557-3570, 10.5194/acp-14-3557-2014, 2014.