

Supplement of Geosci. Model Dev., 10, 721–750, 2017  
<http://www.geosci-model-dev.net/10/721/2017/>  
doi:10.5194/gmd-10-721-2017-supplement  
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*Supplement of*

## **The high-resolution version of TM5-MP for optimized satellite retrievals: description and validation**

**Jason E. Williams et al.**

*Correspondence to:* Jason E. Williams ([williams@knmi.nl](mailto:williams@knmi.nl))

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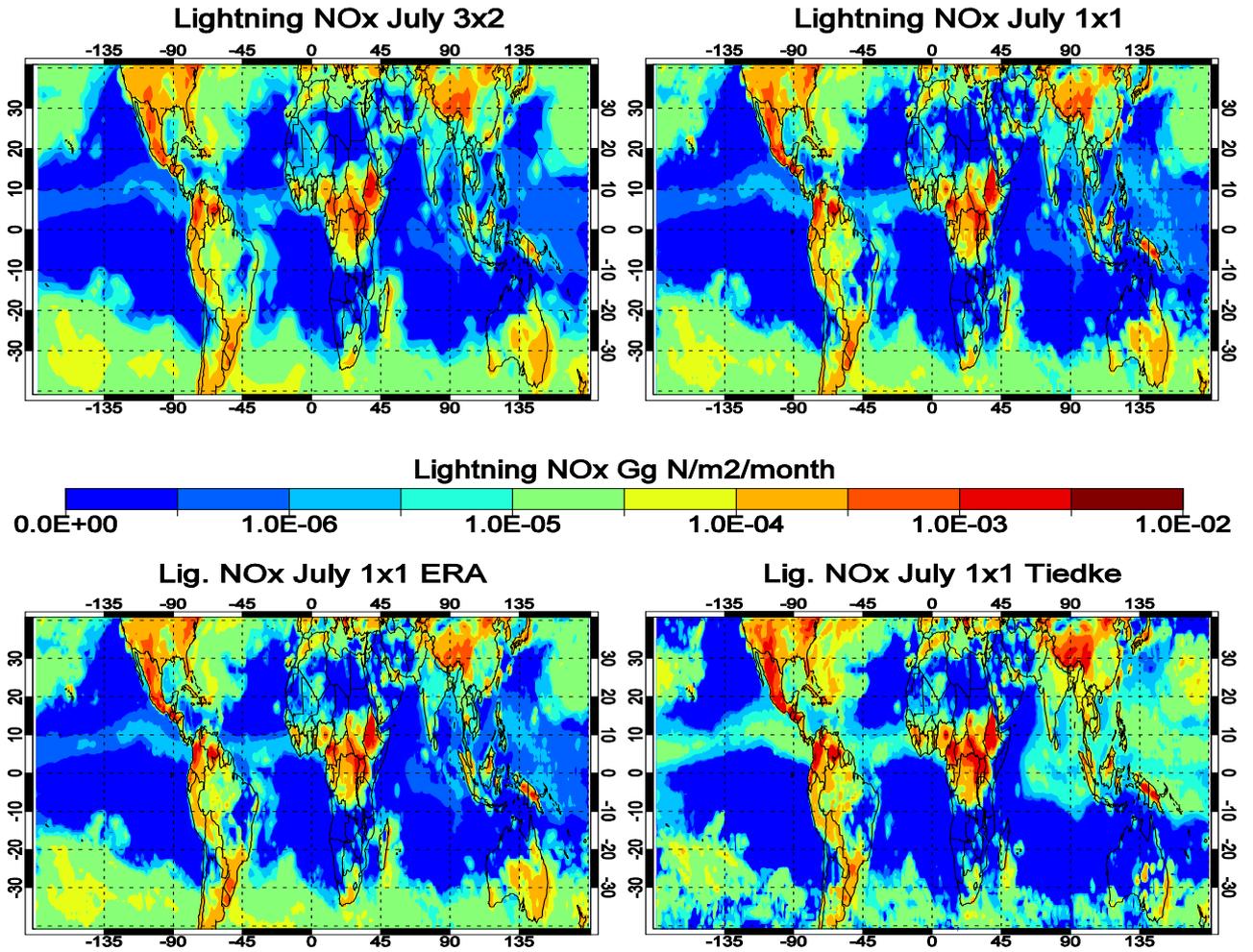


Figure S1a: The global distribution of lightning  $\text{NO}_x$  emissions for July 2006 at  $\sim 400\text{hPa}$  for the  $3^\circ \times 2^\circ$  (top left) and  $1^\circ \times 1^\circ$  (top right) simulations. The corresponding distribution of lightning  $\text{NO}_x$  using the Tiedke (1989) scheme is also shown bottom right. Fluxes are given in  $\text{Gg N/m}^2/\text{month}$ .

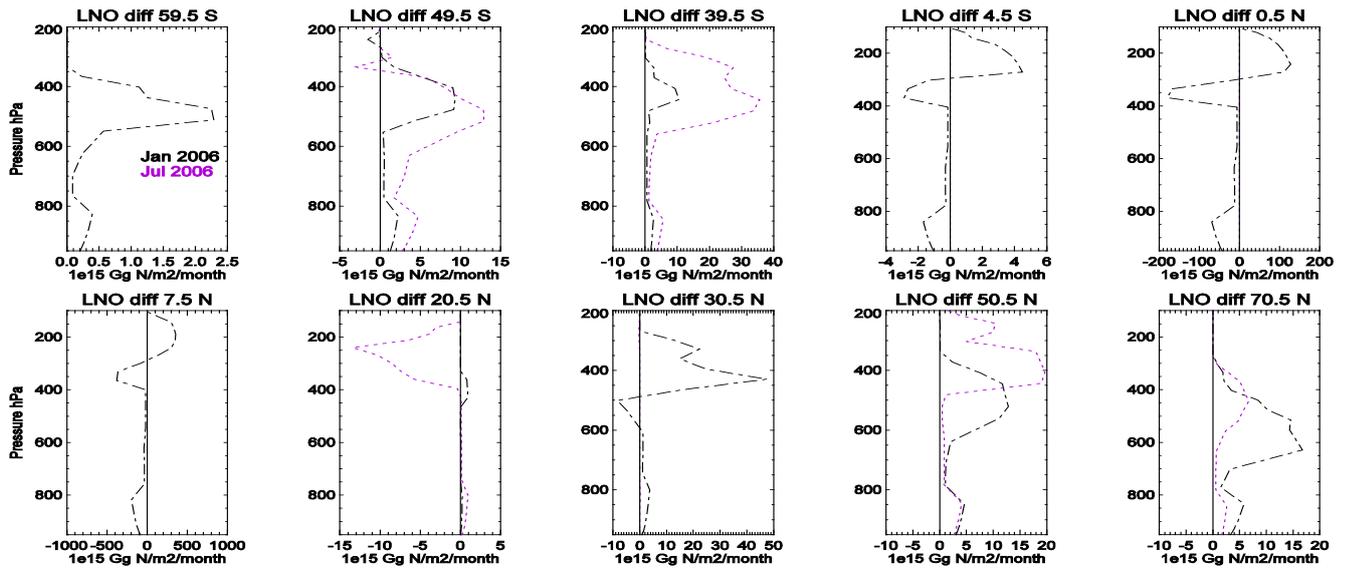


Figure S1b: 1-D profiles of the absolute differences in Lightning  $\text{NO}$  emissions calculated by the parameterization of Meijer et al. (2001) between those calculated using Tiedke and ERA-interim for various latitudes at  $0.5^\circ\text{E}$ . Absolute differences are equal to ERA-Tiedke, and shown for both January and July, being given in  $\text{Gg N m}^2 \text{ month}$  and subsequently scaled up by  $1.0\text{e}15$  for visualization.

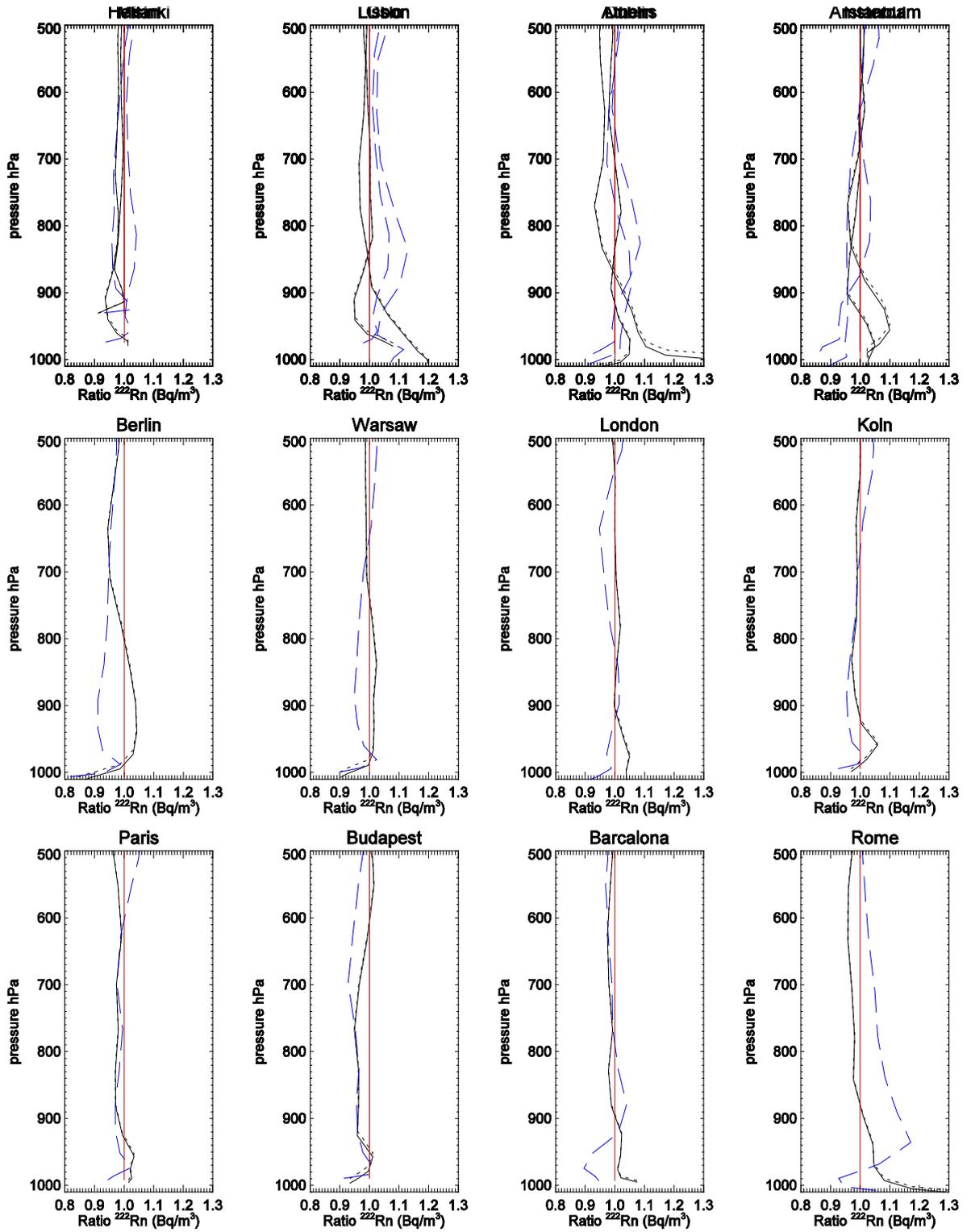


Figure S2: Ratios of the vertical profiles of  $^{222}\text{Rn}$  between  $1^\circ \times 1^\circ / 3^\circ \times 2^\circ$  simulations above selected European cities for January (black) and July (Blue) in 2006. The red line represents the ideal ratio of 1.0 throughout the column.

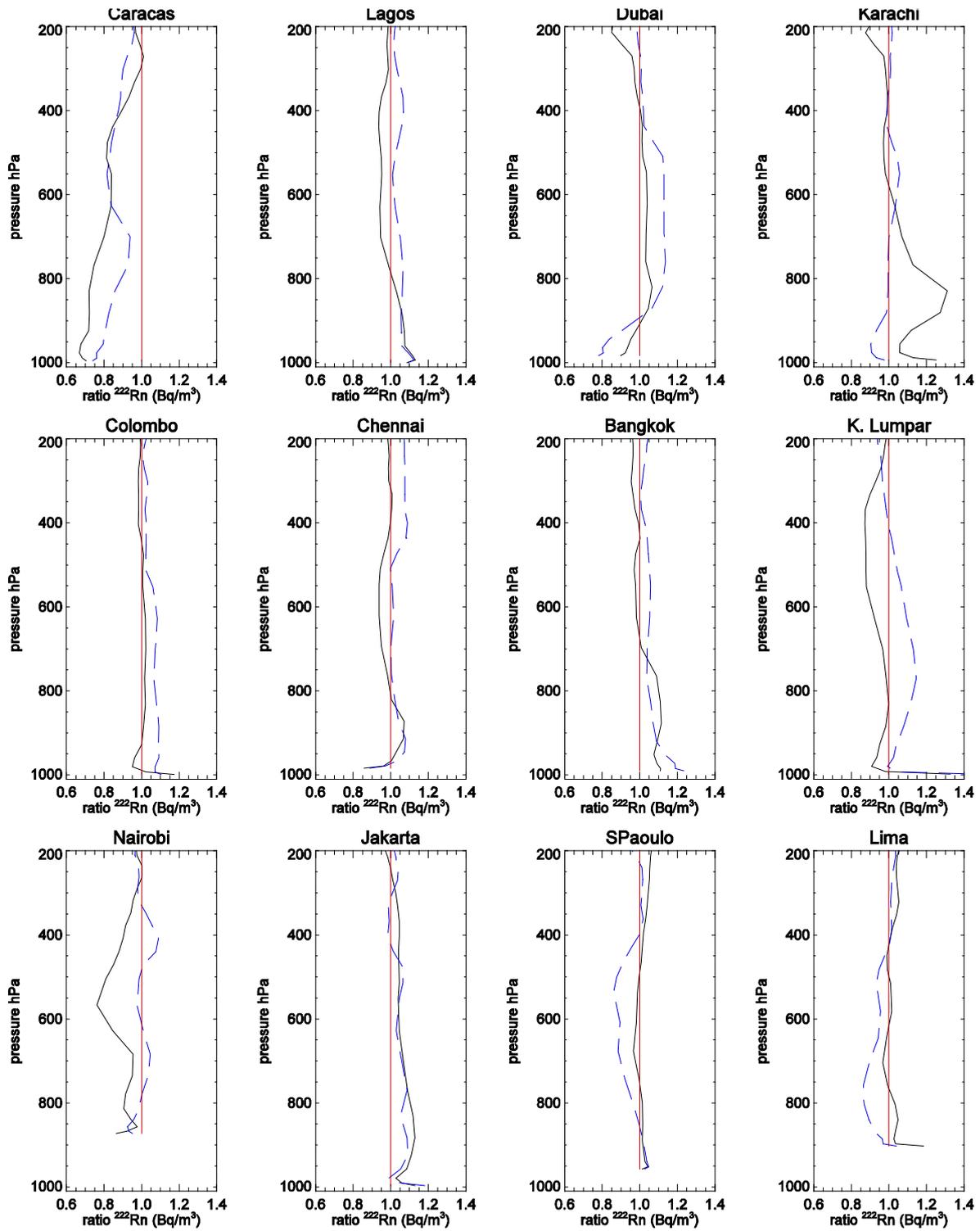


Figure S3: Ratios of the vertical profiles of  $^{222}\text{Rn}$  between  $1^\circ \times 1^\circ / 3^\circ \times 2^\circ$  simulations above selected Tropical cities for January (black) and July (Blue) in 2006. The red line represents the ideal ratio of 1.0 throughout the column.

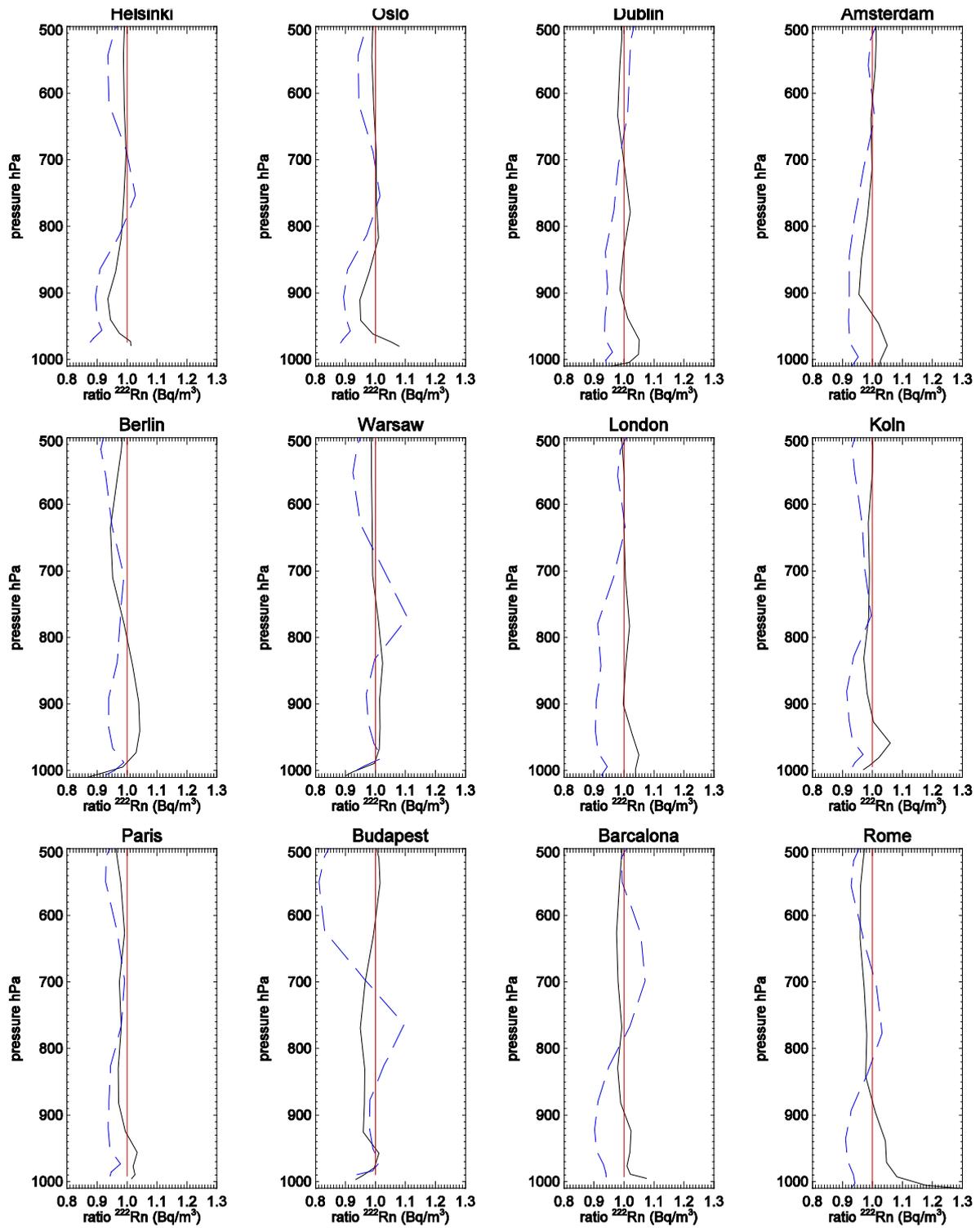


Figure S4: Ratios of the vertical profiles of  $^{222}\text{Rn}$  above selected European cities for January (black) and July (Blue) during 2006. The ratio is representative of  $1^\circ \times 1^\circ / 1^\circ \times 1^\circ$  (Tiedke). The red-line represents the ideal ratio of 1.0 throughout the column.

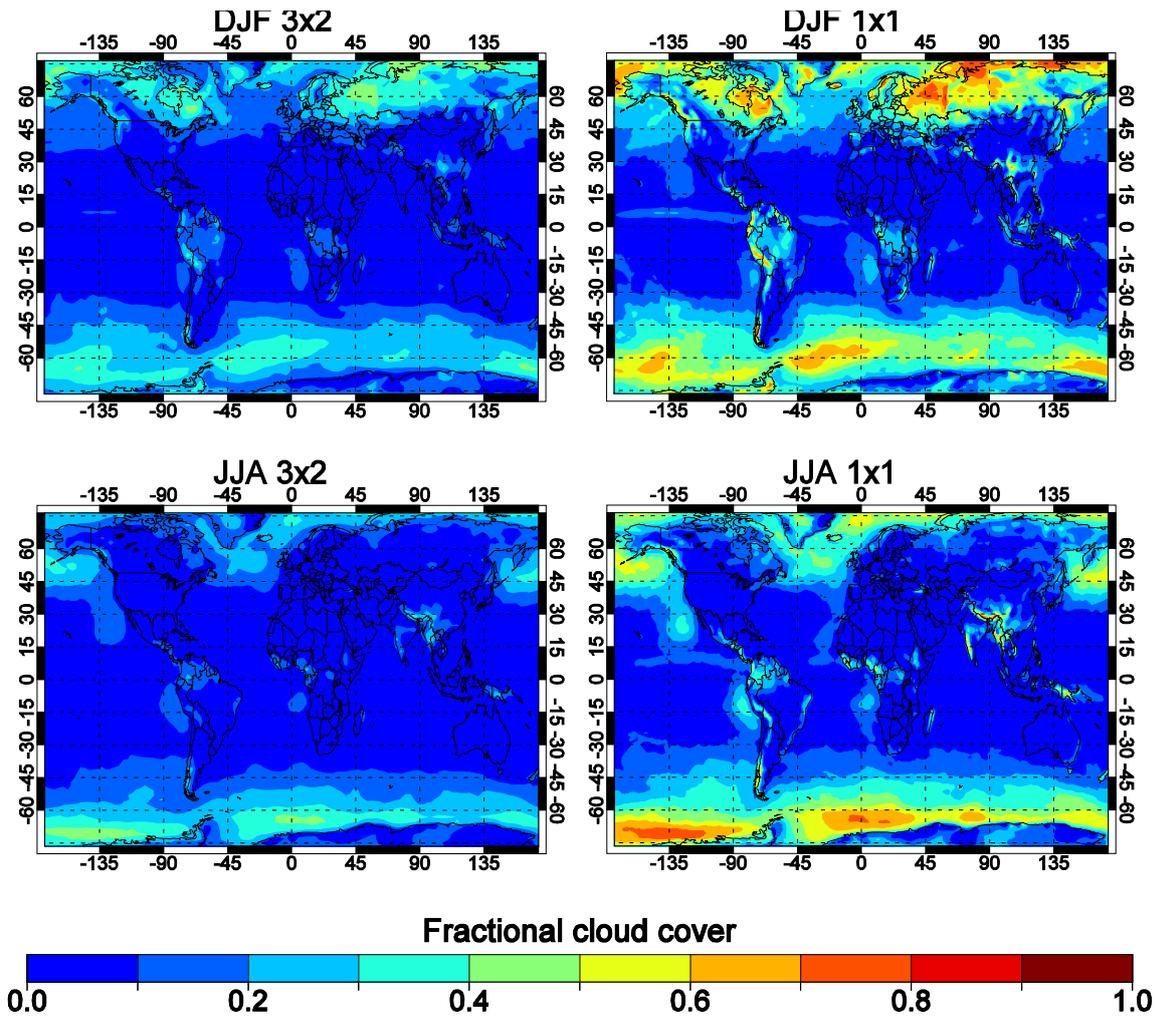


Figure S5: Comparisons of seasonal mean cloud cover values at the top of the boundary layer for (top) DJF and (bottom) JJA between the 3° x 2° (left) and 1° x 1° (right) simulations. The values shown are representative of 1-2 km of the troposphere, which exhibits high cloud incidence.

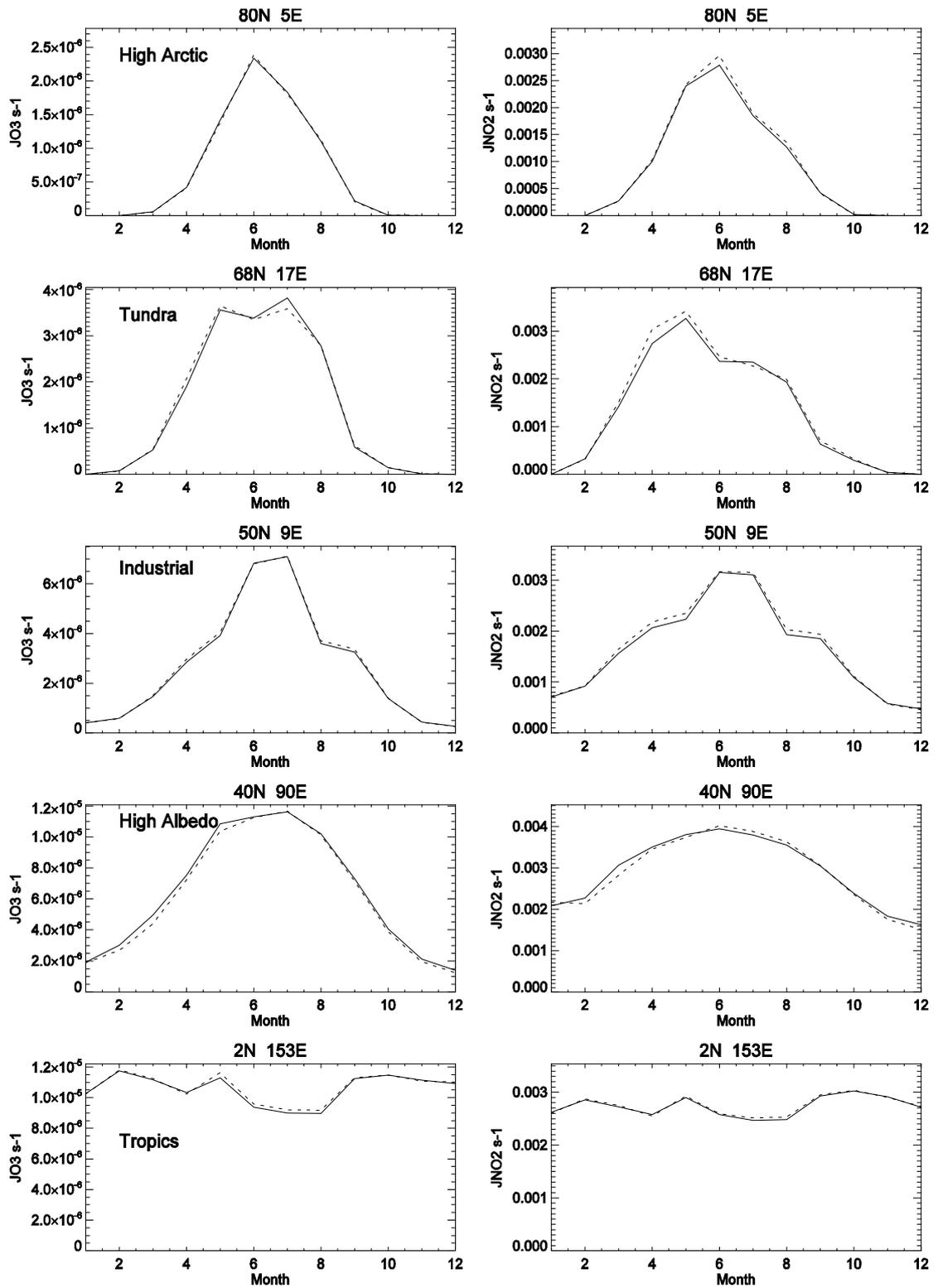


Figure S6: Comparisons of monthly mean  $J_{O_3}$  (left) and  $J_{NO_2}$  (right) values at the surface between the  $3^\circ \times 2^\circ$  (solid line) and  $1^\circ \times 1^\circ$  (dashed line) simulations. The locations selected are identical to those shown in Williams et al. (2012), where the type of region is given in each panel to the left.

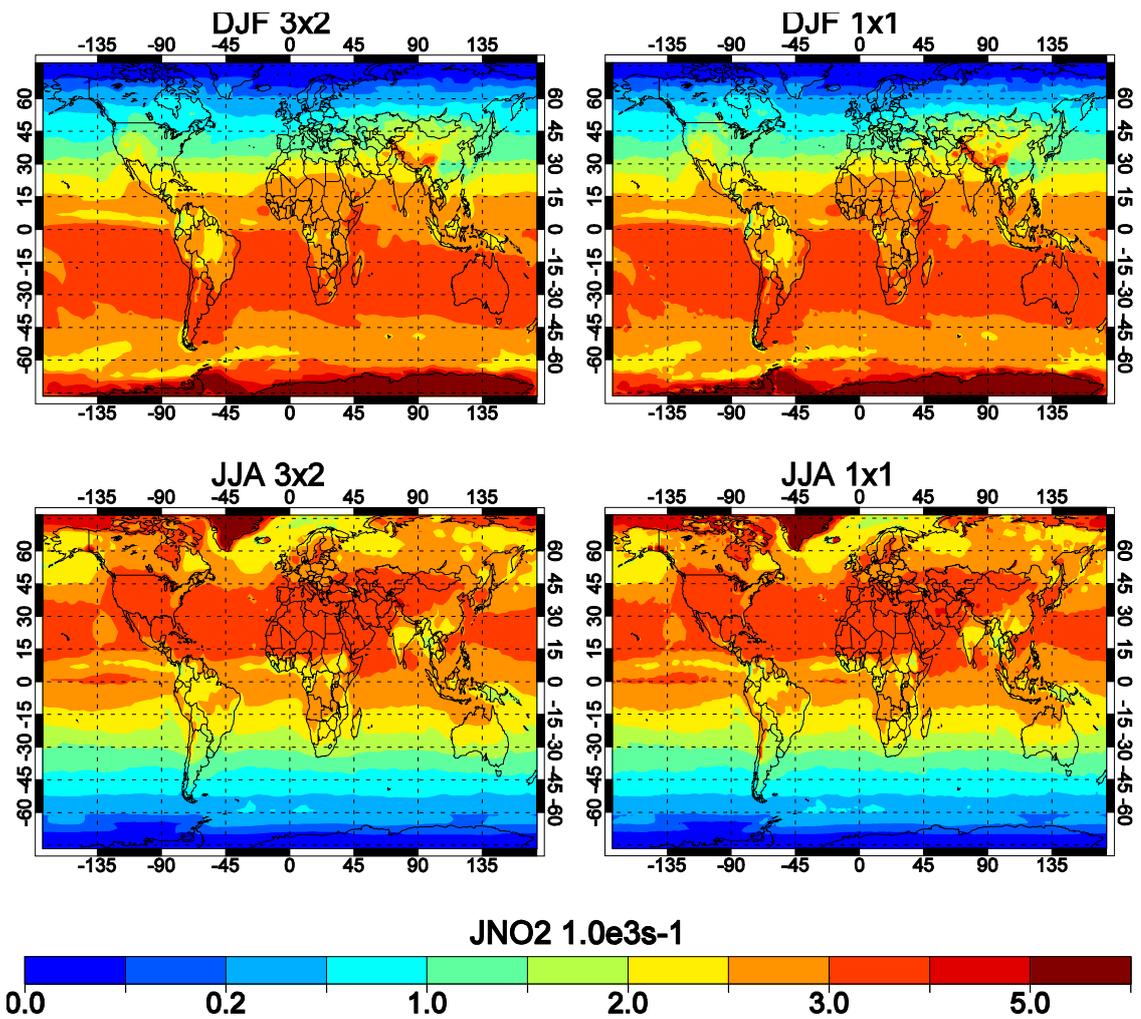


Figure S7: Comparisons of near-surface seasonal mean  $J_{NO_2}$  values for (top) DJF and (bottom) JJA between the  $3^\circ \times 2^\circ$  (left) and  $1^\circ \times 1^\circ$  (right) simulations. The values shown are representative of the lowest km of the troposphere.

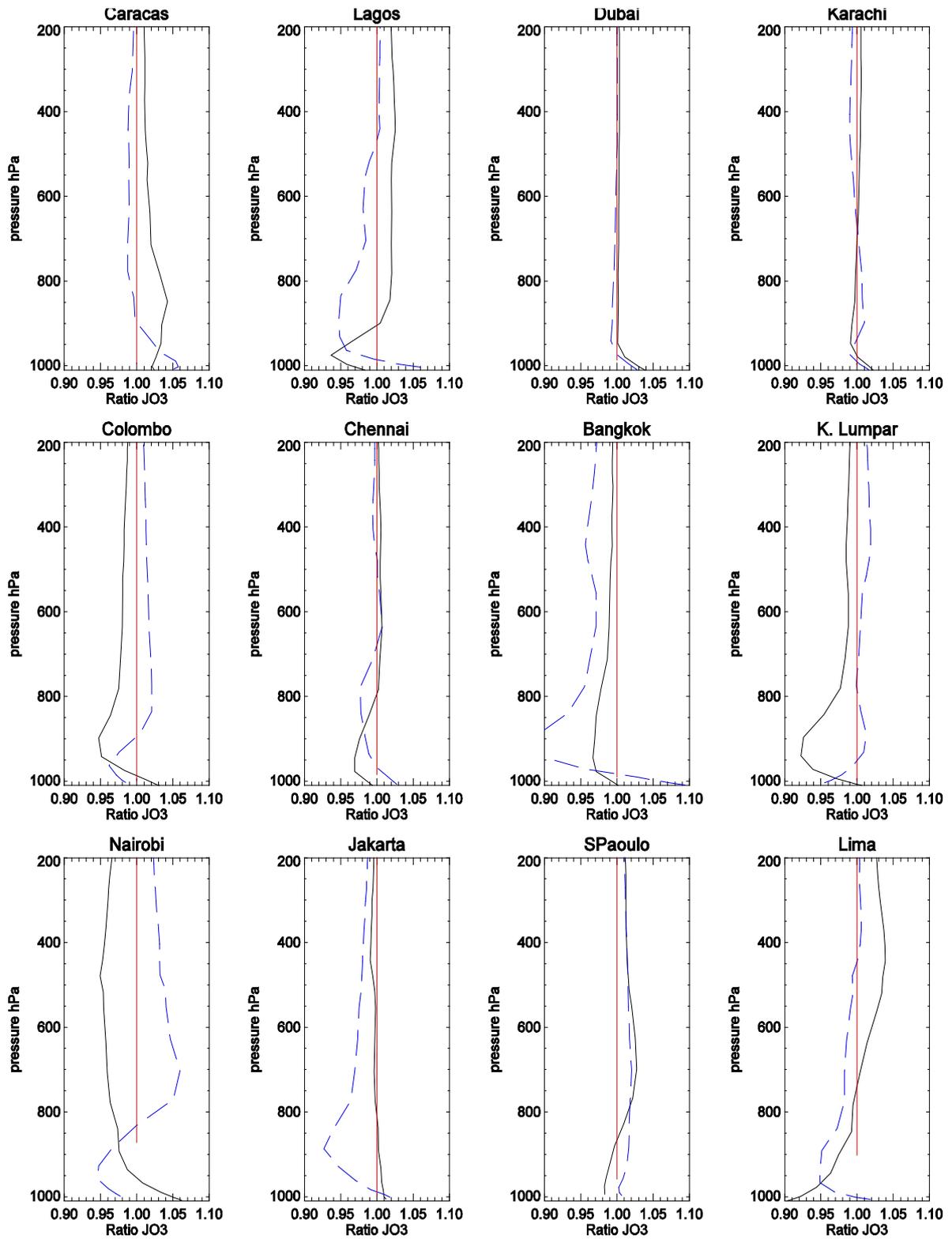


Figure S8a: Comparisons of the ratio of monthly mean  $J_{O_3}$  profiles above selected tropical cities for January (black) and July (Blue) in 2006. The ratio is equal to  $1^\circ \times 1^\circ / 3^\circ \times 2^\circ$  values.

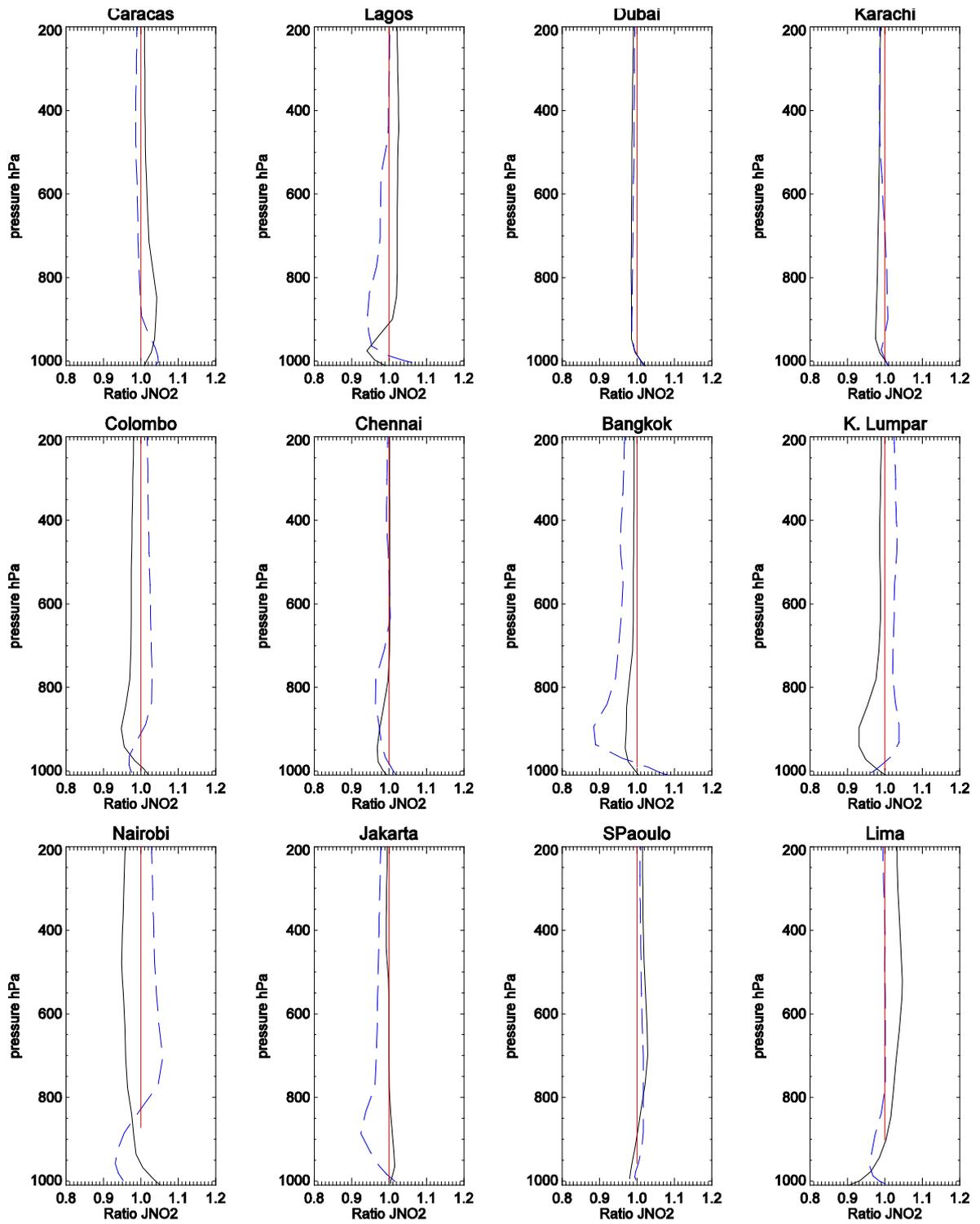


Figure S8b: Comparisons of the ratio of monthly mean  $J_{NO_2}$  profiles above selected tropical cities for January (black) and July (Blue) in 2006. The ratio is equal to  $1^\circ \times 1^\circ / 3^\circ \times 2^\circ$  values.

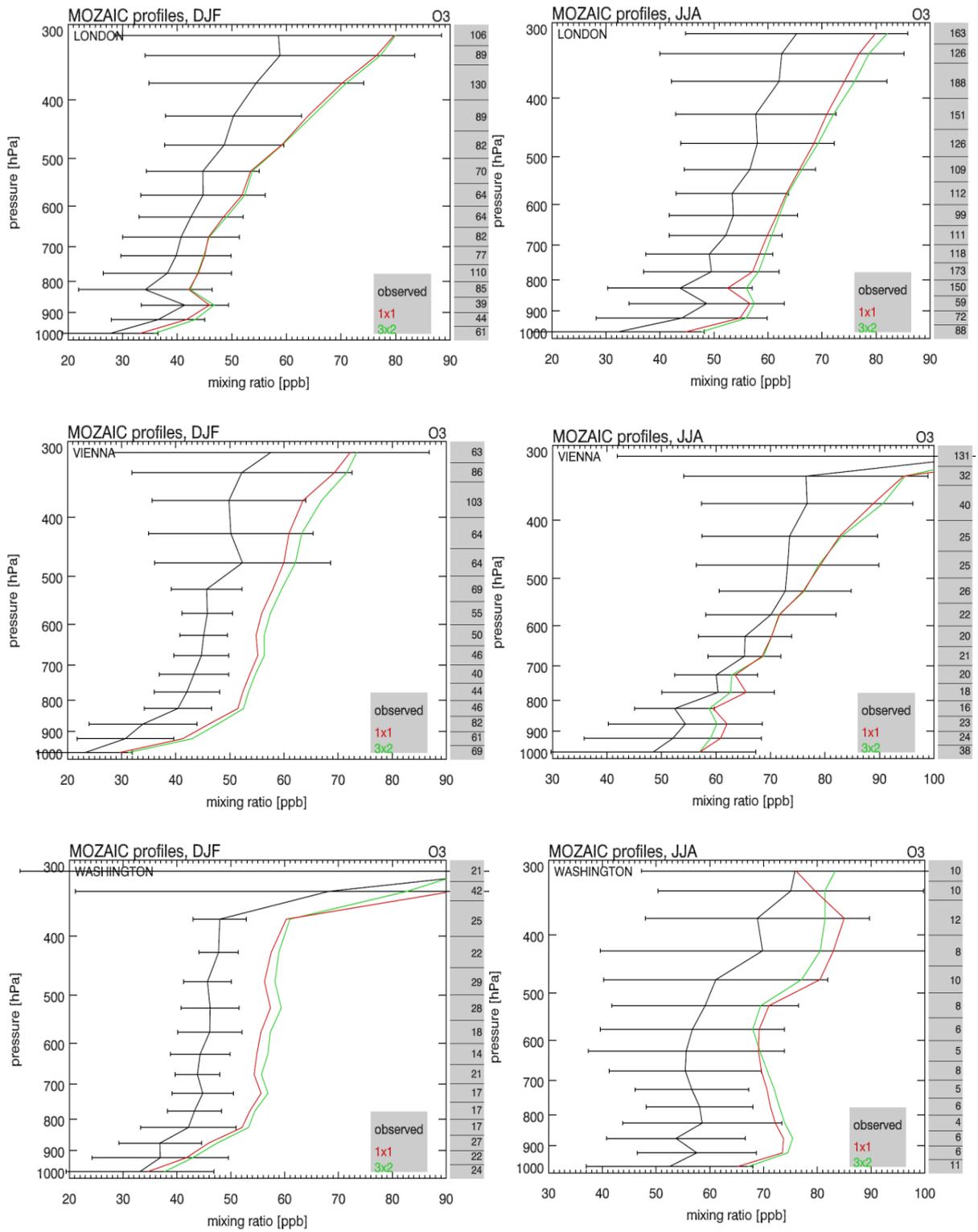


Figure S9a: Seasonal comparisons of tropospheric O<sub>3</sub> profiles (ppb) taken as part of the MOZIAC flight program for (left) DJF and (right) JJA for (top) London, (middle) Vienna and (bottom) Washington.

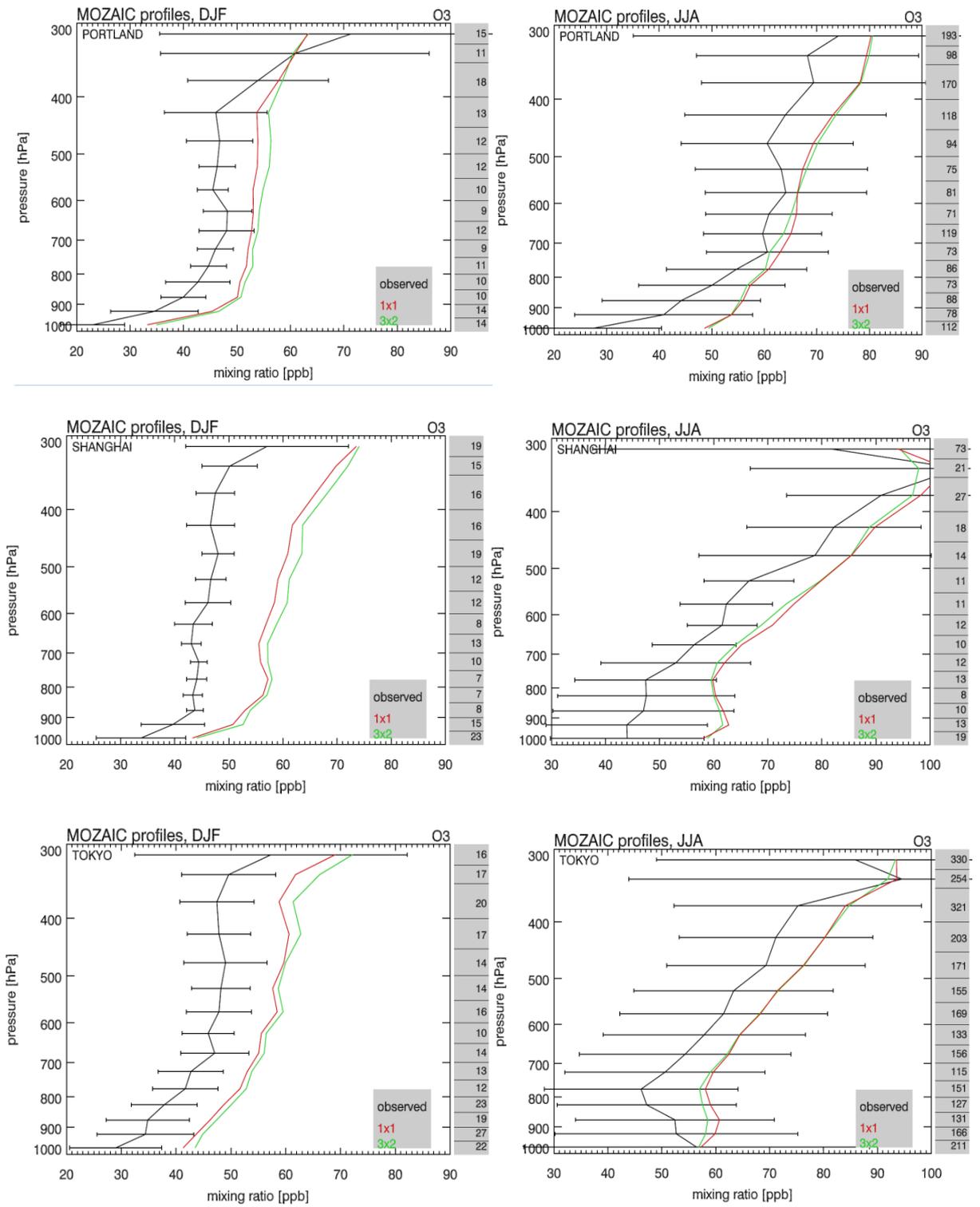


Figure S9b: As for Fig. S9a except for Portland (top), Shanghai (middle) and Tokyo (bottom).

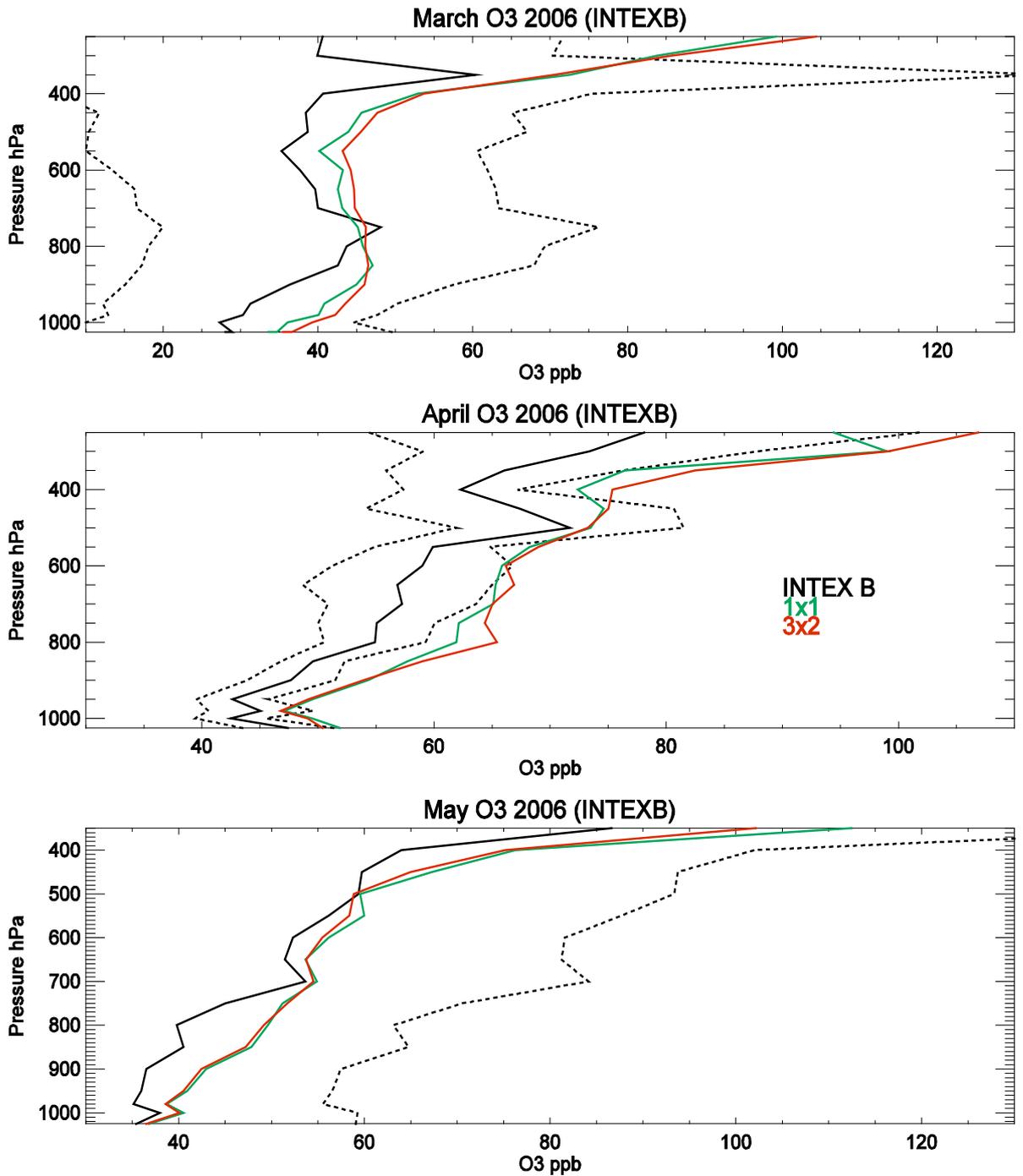


Figure S10: Comparisons of monthly composites of tropospheric O<sub>3</sub> profiles (ppb) against measurements taken during the INTEX-B campaign between March and May 2006 for both 3° x 2° and 1° x 1° simulations. The dotted line represents the 1-σ variability associated with the measurements.

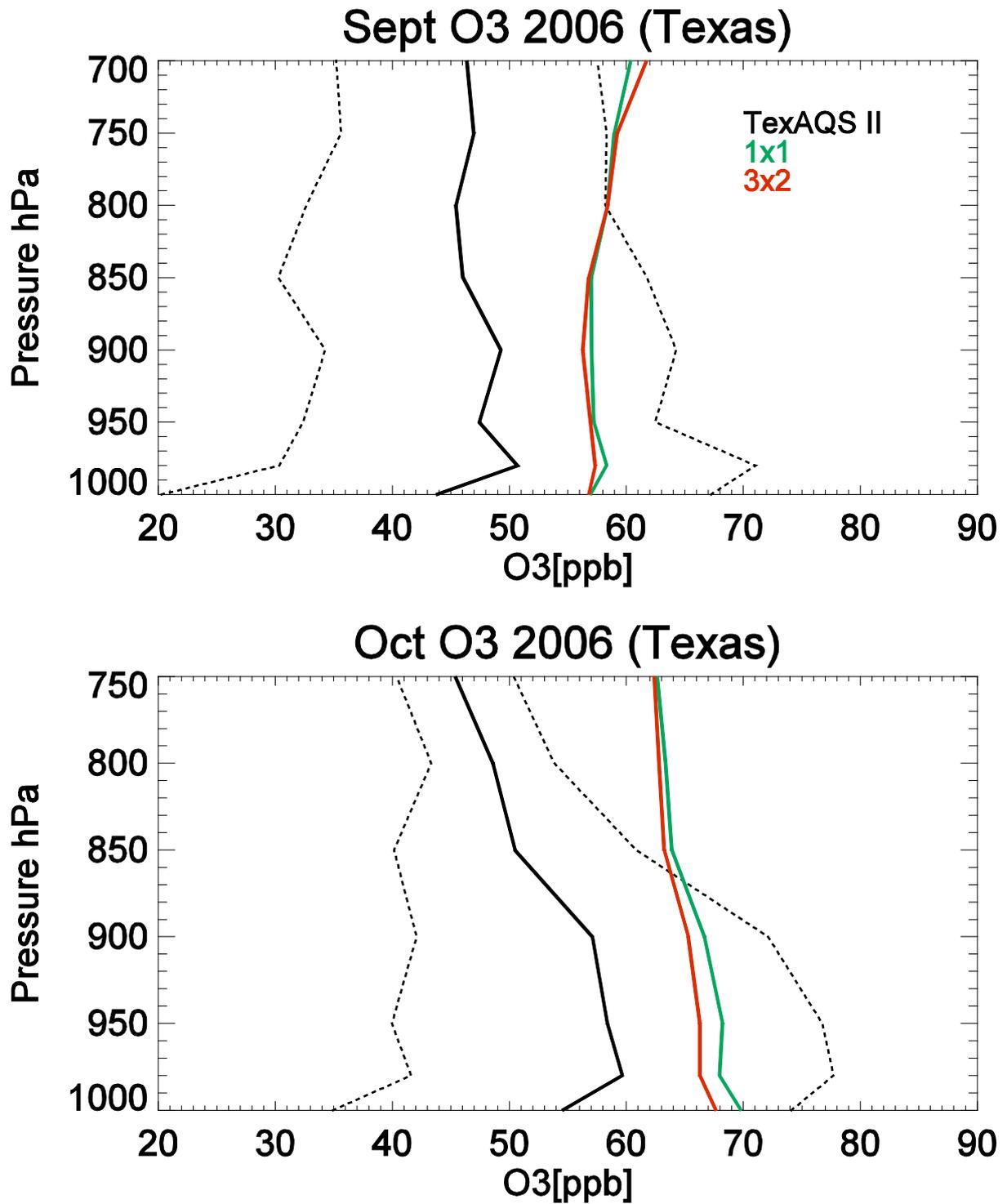


Figure S11: Comparisons of monthly tropospheric O<sub>3</sub> profiles assembled from data taken during September and October 2006 as part of the Texas-AQS measurement campaign. The dotted line represents the 1- $\sigma$  variability associated with the measurements. For details of the flight paths the reader is referred to the details given in Parrish et al. (2009).

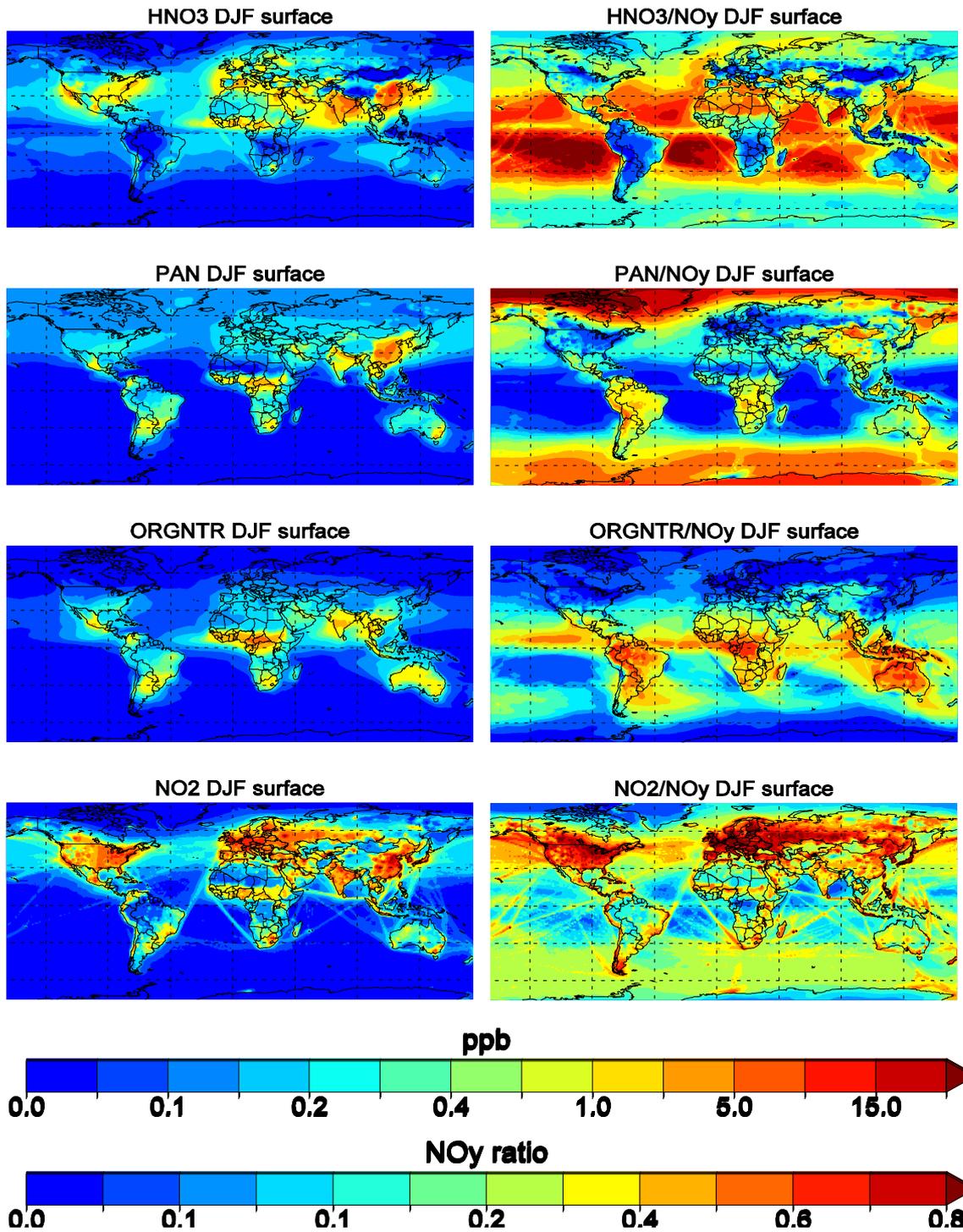


Figure S12: The horizontal mean distribution of (top to bottom) HNO<sub>3</sub>, PAN, ORGNTR and NO<sub>2</sub> in the 1° x 1° simulation for DJF, along with the corresponding NO<sub>y</sub> ratios.

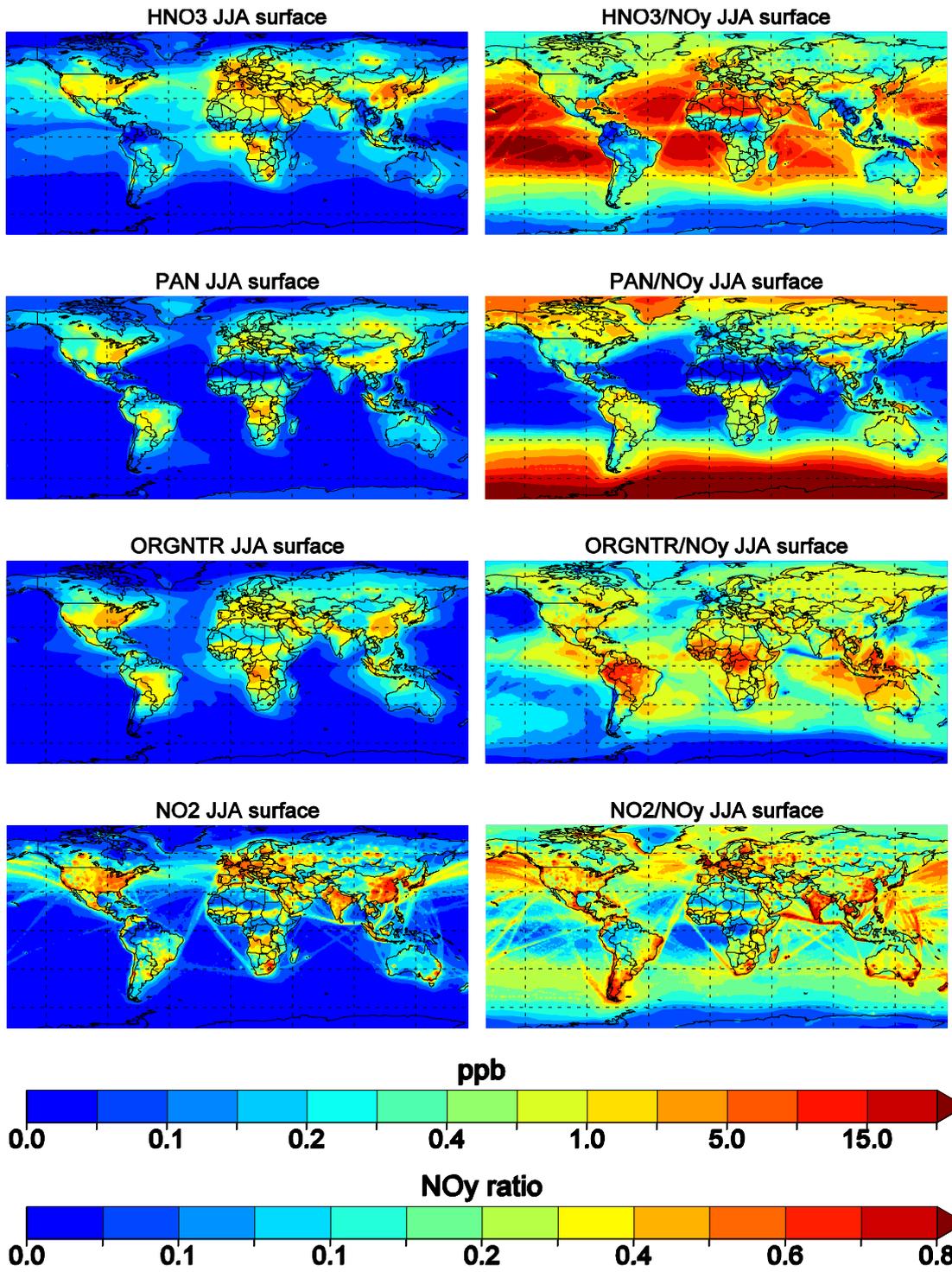


Figure S13: As for Fig S12 except season JJA.

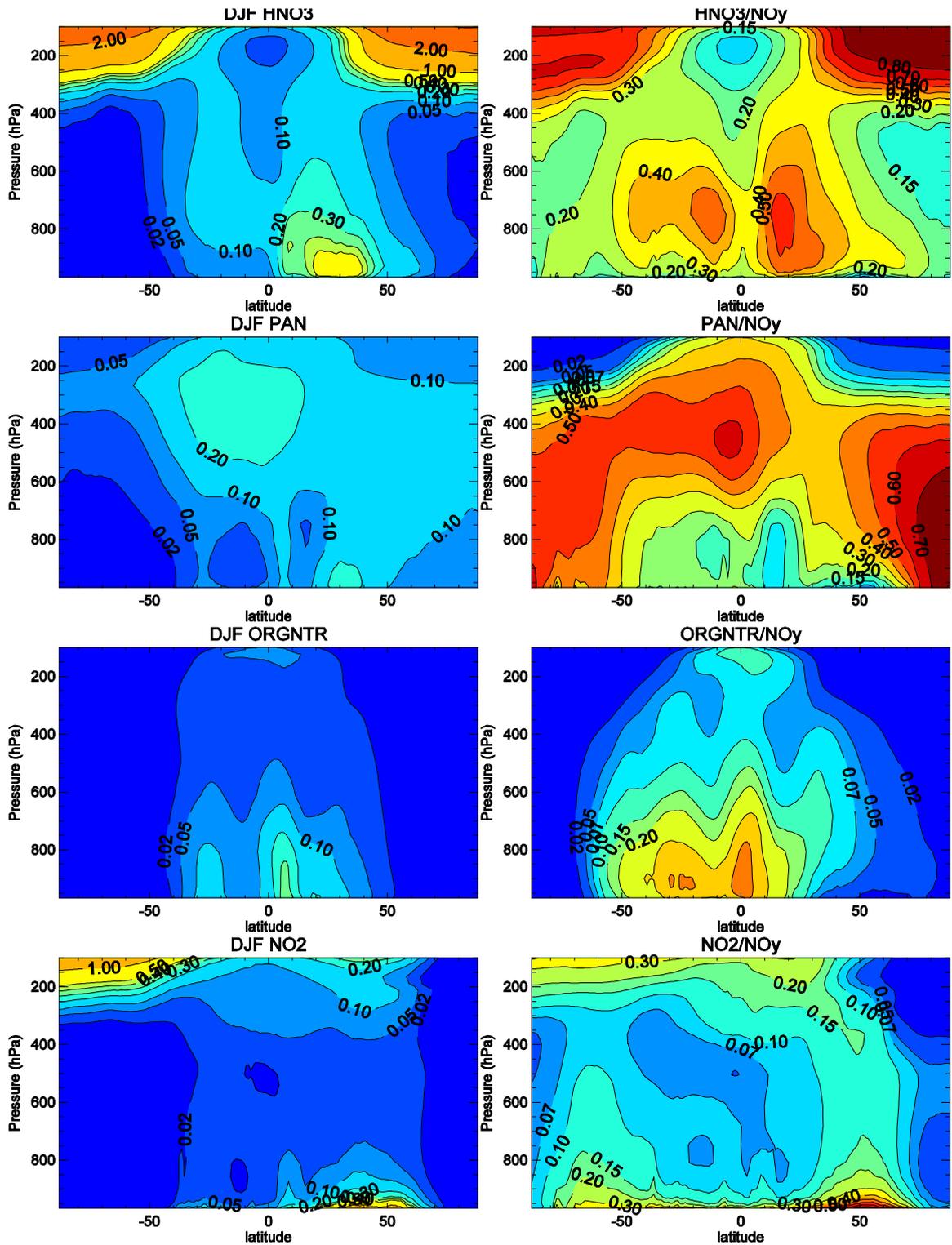


Figure S14: The seasonal zonal mean distribution of (top to bottom) in the  $1^\circ \times 1^\circ$  simulation for  $\text{HNO}_3$ , PAN, ORGNTR and  $\text{NO}_2$  for DJF with the corresponding  $\text{NO}_y$  ratios. The ratio is calculated as  $[\text{species}]/[\text{NO}_y]$ , where a definition of  $\text{NO}_y$  is given in the text.

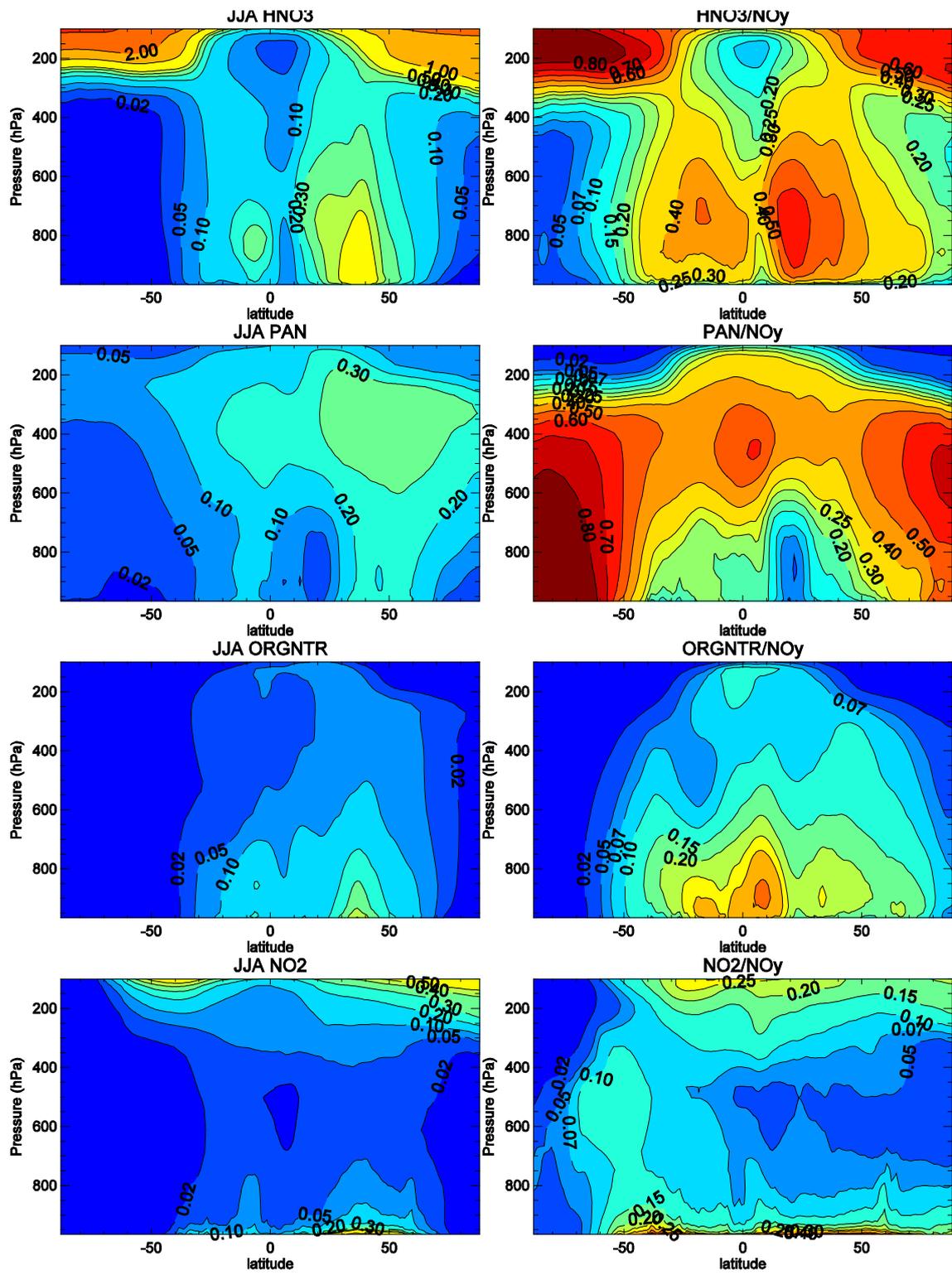


Figure S15: As for Fig. S14 except for season JJA.

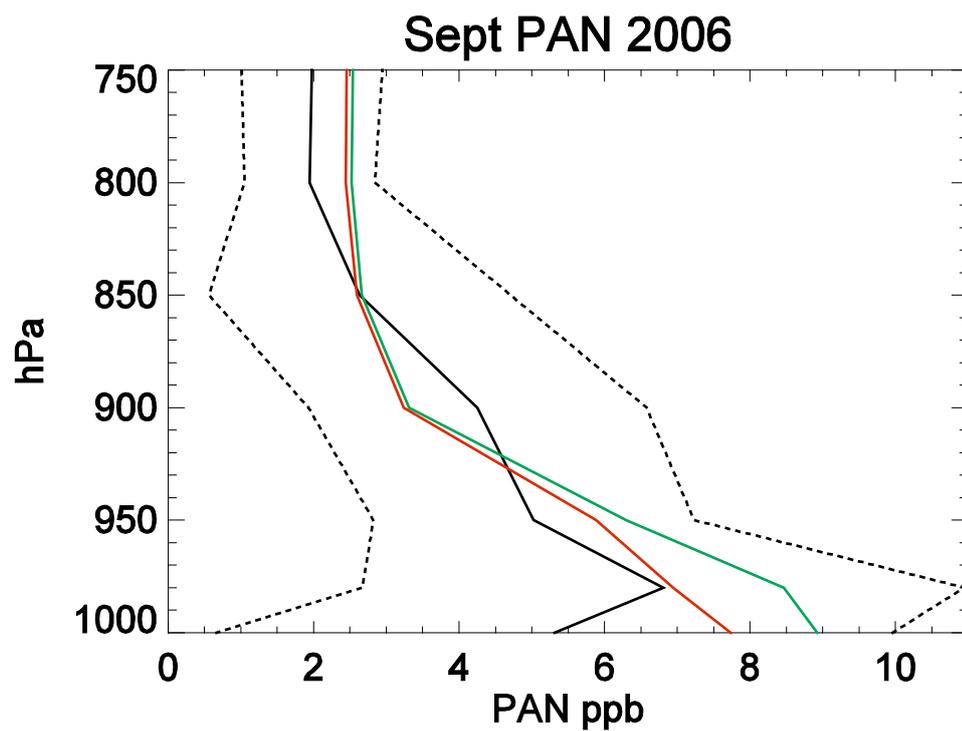
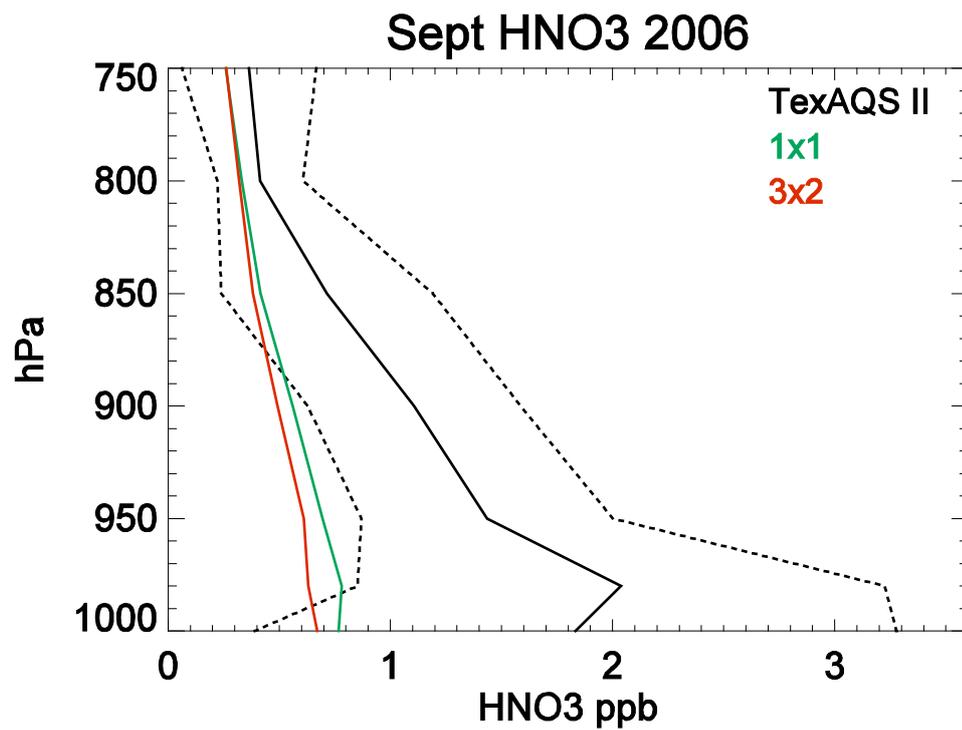


Figure S16: Comparisons of monthly tropospheric HNO<sub>3</sub> (top) and PAN (bottom) profiles during September 2006 above Texas. The 1- $\sigma$  deviation from the measurements is shown as the dotted line for each species. For details of the flight paths the reader is referred Parrish et al. (2009).

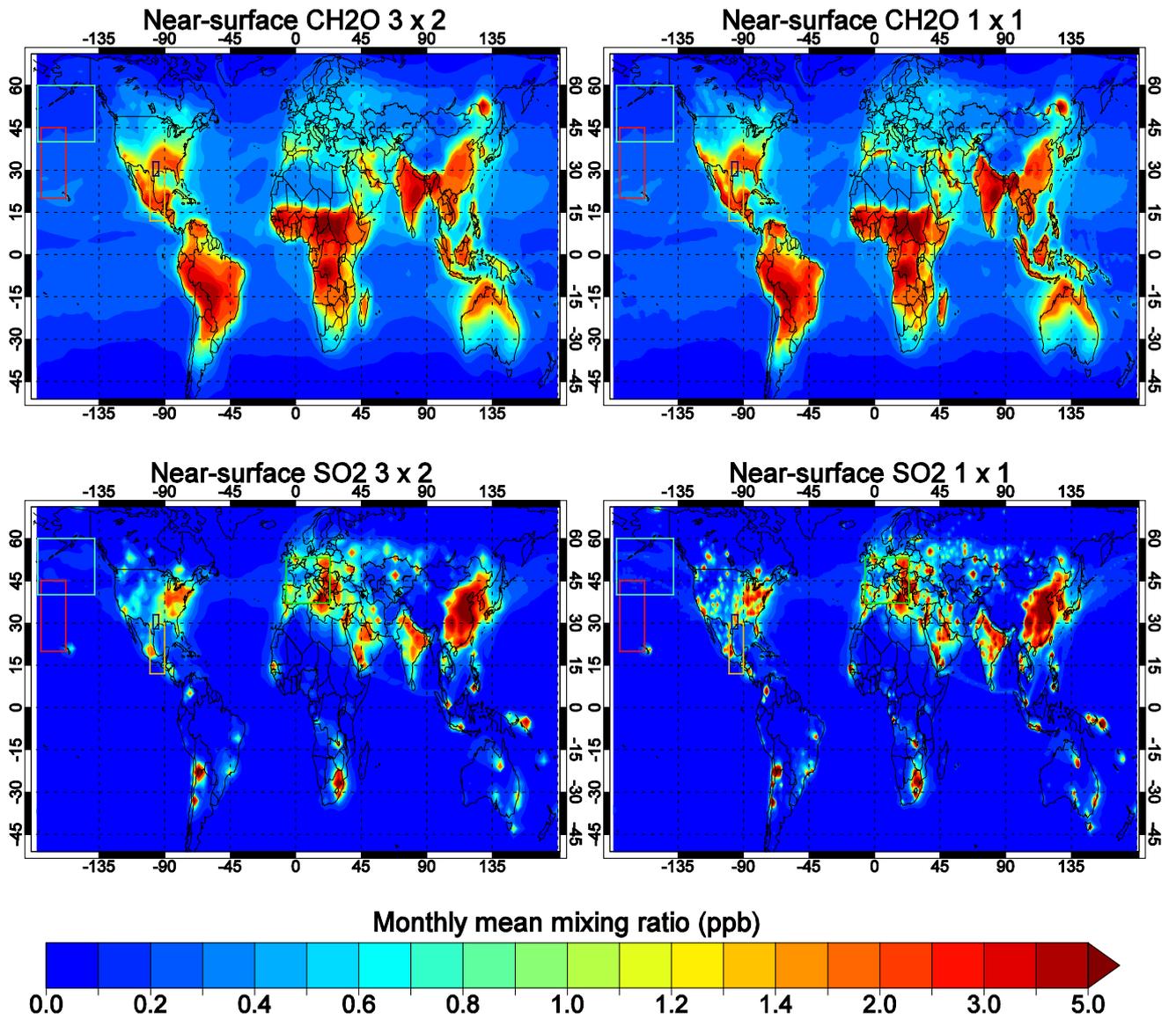


Figure S17: The near-surface distribution in tropospheric  $\text{CH}_2\text{O}$  (top) and  $\text{SO}_2$  (bottom) for May 2006 from the  $3^\circ \times 2^\circ$  (left) and  $1^\circ \times 1^\circ$  (right) TMS-MP simulations. Also shown are the locations of the INTEXB and Texas-AQSII measurement campaigns, and the extent of the EMEP network in the European domain for  $\text{SO}_2$  comparisons.

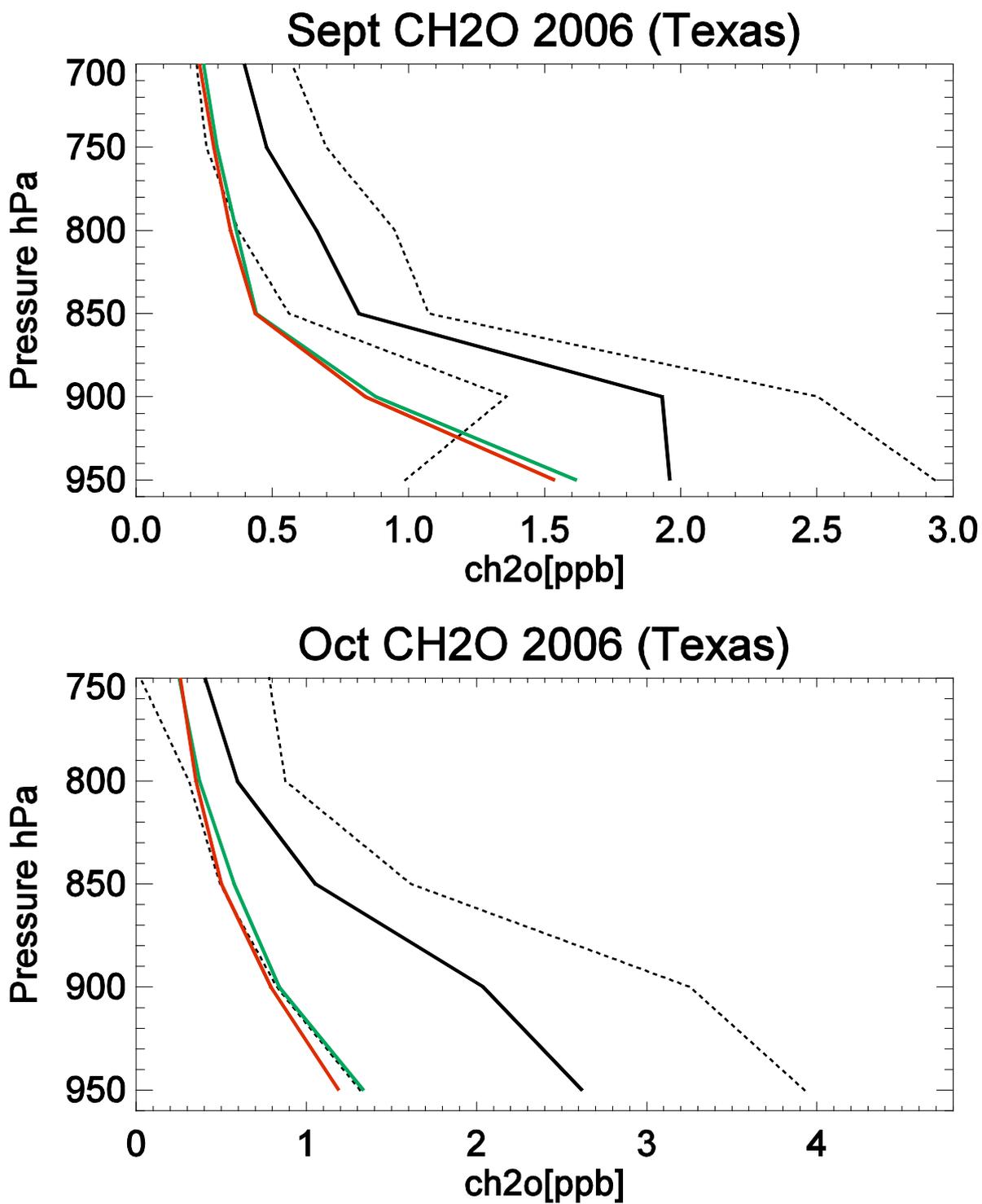


Figure S18: Comparisons of monthly tropospheric CH<sub>2</sub>O profiles assembled from data taken during September and October 2006 as part of the Texas-AQS measurement campaign. The 1- $\sigma$  deviation from the measurements is shown as the dotted line. For details of the flight paths the reader is referred to the details given in Parrish et al. (2009).

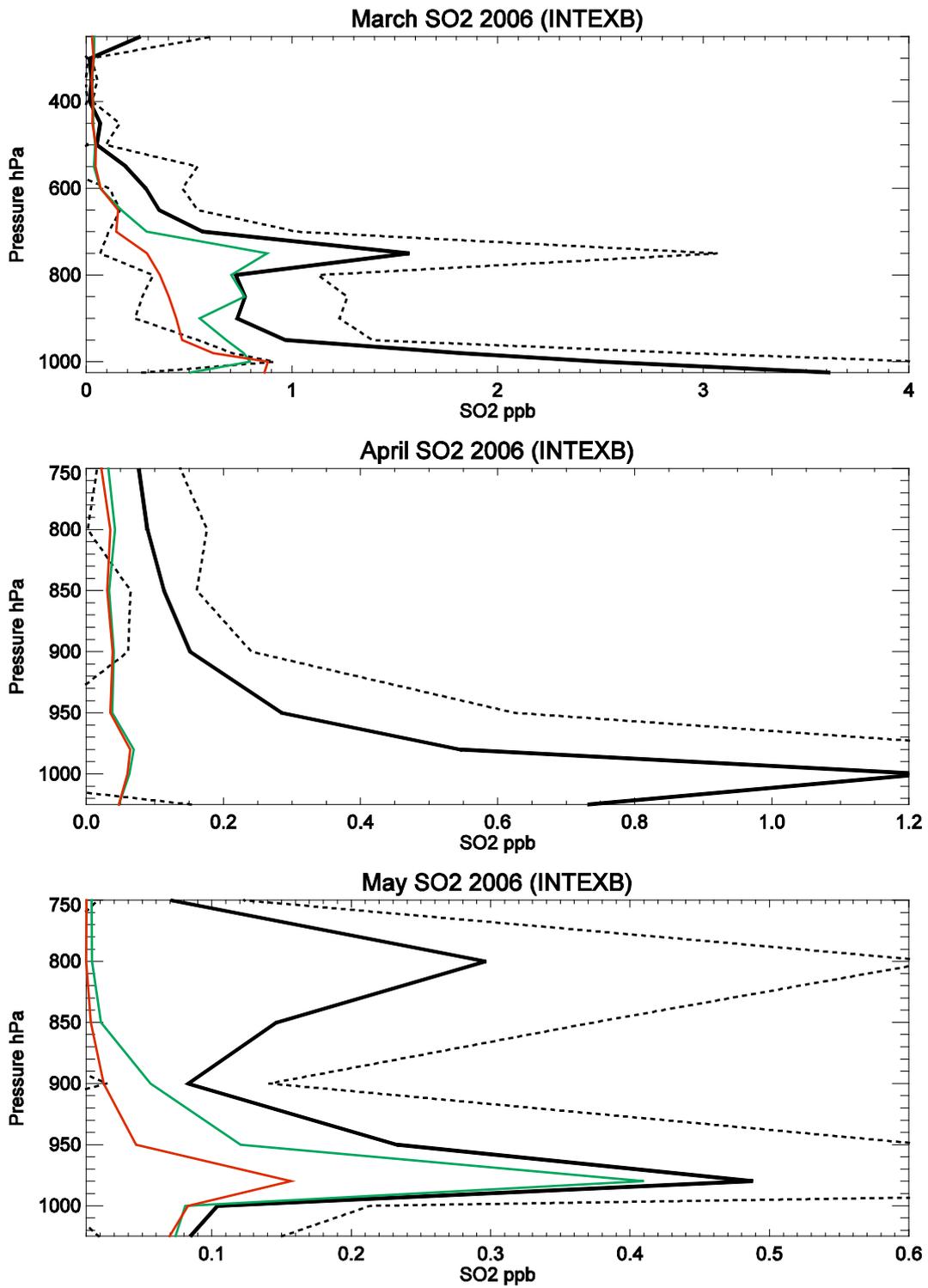


Figure S19: Comparisons of the vertical distribution of SO<sub>2</sub> from both 3°x2° and 1°x1° simulations against measurements made as part of the INTEX B campaign during 2006. The 1- $\sigma$  deviation from the measurements is shown as the dotted line for each species. For details on the exact location of the flights the reader is referred to Parrish et al. (2009).

Table S1: The tropospheric chemical budget terms for the chemical production (CP), chemical destruction (CD) and accumulated deposition for HNO<sub>3</sub>, PAN and ORGNTR given in Tg N yr<sup>-1</sup> for the 1° x 1° simulation during 2006. Loss of HNO<sub>3</sub> into NO<sub>3</sub><sup>-</sup> accounts for the missing HNO<sub>3</sub> loss term. The chemical troposphere is defined according to Stevenson et al. (2006) and fixed across simulations. Percentage differences are given in the parenthesis when compared against the corresponding budget terms from the 3° x 2° simulation (1° x 1°/3° x 2°). The SH, Tropics and NH are defined as 30-90°S, 30°S-30°N and 30-90°N, respectively.

Budget Term (Tg/N)	Global	SH	Tropics	NH
Strat. Nudge	0.3	0.1	0.1	0.1
HNO <sub>3</sub> CP	44.0 (0.3)	1.9 (1.0)	22.8 (-1.0)	19.3 (1.7)
HNO <sub>3</sub> CD	6.7 (-4.8)	0.5 (4.7)	3.9 (-2.5)	2.3 (6.7)
HNO <sub>3</sub> Dep.	35.9 (1.5)	2.0 (-0.5)	18.2 (1.6)	15.6 (1.7)
PAN CP	199.1 (-2.4)	7.6 (-3.0)	150.8 (-2.7)	40.7 (-1.1)
PAN CD	197.5 (-2.4)	7.7 (-3.0)	150.2 (-2.7)	39.7 (-1.1)
PAN Dep.	1.6 (2.5)	0.1 (1.0)	0.7 (-0.5)	0.8 (-4.6)
ORGNTR CP	9.8 (-4.8)	0.3 (-3.0)	6.9 (-5.5)	2.6 (-3.4)
ORGNTR CD	4.2 (-4.6)	0.2 (-5.0)	2.9 (-3.4)	1.3 (-0.8)
ORGNTR Dep.	5.7 (-3.6)	0.5 (-2.0)	3.5 (-4.4)	1.8 (-2.8)

Table S2: The tropospheric chemical budget terms for the short-lived N-species HONO, HNO<sub>4</sub> and N<sub>2</sub>O<sub>5</sub> (Tg N yr<sup>-1</sup>) during 2006 for the 1° x 1° simulation. Both HNO<sub>4</sub> and N<sub>2</sub>O<sub>5</sub> exist in chemical equilibrium with their respective chemical precursors which accounts for the dominant loss terms (not given), where only oxidation by OH and heterogeneous conversion terms are provided. The chemical troposphere is defined according to Stevenson et al. (2006) and fixed across simulations. Percentage differences are given in the parenthesis when compared against the corresponding budget terms from the 3° x 2° simulation (1° x 1°/3° x 2°). The SH, Tropics and NH are defined as 30-90°S, 30°S-30°N and 30-90°N, respectively.

Budget Term (Tg N yr <sup>-1</sup> )	Global	SH	Tropics	NH
HO + NO	22.9 (-3.0)	1.3 (-0.8)	16.4 (-6.4)	5.2 (9.0)
OH + HONO	2.6 (-1.9)	0.1 (-)	1.8 (-5.2)	0.7 (6.3)
HNO <sub>4</sub> CP	168.7 (-1.4)	7.5 (-0.4)	106.4 (-1.2)	54.7 (-1.7)
OH + HNO <sub>4</sub>	13.5 (-1.2)	1.1 (0.9)	8.4 (-2.0)	4.0 (-)
N <sub>2</sub> O <sub>5</sub> CP	628.4 (12.4)	11.2 (24.9)	322 (12.8)	226 (11.6)
N <sub>2</sub> O <sub>5</sub> + aero	6.3 (5.9)	0.1 (14.3)	1.7 (12.0)	4.5 (3.9)
N <sub>2</sub> O <sub>5</sub> + cloud	3.2 (-1.9)	0.1 (-)	0.7 (4.8)	2.5 (0.8)
N <sub>2</sub> O <sub>5</sub> Dep.	0.3 (-3.0)	-	0.1 (-)	0.2 (-7.5)