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Supplementary Materials for

Aircraft observations since the 1990s reveal increases of tropospheric ozone at multiple locations across the Northern Hemisphere

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I- Tables and Figures

Table S1a. Trends in median ozone above the six mid-latitude and subtropical regions throughout the troposphere. *Trend values and associated uncertainties are derived from a linear regression of median ozone values between 1994 and 2016 for the IAGOS flights and between 2004 and 2019 for the OMI/MLS tropospheric column ozone (TCO). For each region trends are reported for annual data (all months of the year) and for the cold (October-March) and warm (April-September) seasons. The absolute change of ozone over the 22-year period is reported, as is the linear trend in units of nmol mol^{-1} decade^{-1} and % decade^{-1}. Bold font indicates p-values ≤ 0.05, while italicized font indicates p-values in the range 0.05 – 0.10.*

	Pressure level	Annual		Cold season		Warm Season	
		nmol mol^{-1} [%]	nmol mol^{-1} decade^{-1} [% decade^{-1}]	nmol mol^{-1} [%]	nmol mol^{-1} decade^{-1} [% decade^{-1}]	nmol mol^{-1} [%]	nmol mol^{-1} decade^{-1} [% decade^{-1}]
Europe 16997 flights for 1994-2004 6494 flights for 2011-2016 28905 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**2.8 [5.6]**	**1.3 ± 0.2 p<0.01 [2.5]**	**3.7 [7.3]**	**1.7 ± 0.2 p<0.01 [3.2]**	**1.7 [3.4]**	**0.8 ± 0.4 p<0.01 [1.5]**
	Free troposphere (700-300 hPa)	**2.8 [5.2]**	**1.3 ± 0.2 p<0.01 [2.4]**	**3.7 [6.8]**	**1.7 ± 0.3 p<0.01 [3.1]**	**1.9 [3.5]**	**0.9 ± 0.4 p<0.01 [1.6]**
	Upper troposphere (400-300 hPa)	**3.9 [6.8]**	**1.8 ± 0.4 p<0.01 [3.1]**	**5.0 [8.6]**	**2.3 ± 0.5 p<0.01 [3.9]**	**2.7 [4.6]**	**1.2 ± 0.6 p<0.01 [2.1]**
	Middle troposphere (650 hPa)	**2.5 [4.8]**	**1.1 ± 0.3 p<0.01 [2.2]**	**3.1 [6.0]**	**1.4 ± 0.3 p<0.01 [2.7]**	**1.5 [2.8]**	**0.7 ± 0.4 p<0.01 [1.3]**
	Lower tropopshere (950-800 hPa)	**2.5 [6.3]**	**1.1 ± 0.3 p<0.01 [2.9]**	**3.3 [8.5]**	**1.5 ± 0.3 p<0.01 [3.9]**	**1.4 [3.5]**	**0.6 ± 0.4 p=0.01 [1.6]**
	OMI/MLS (TCO)	**2.7 [6.2]**	**1.9 ± 1.7 p=0.03 [3.8]**	**4.1 [6.9]**	**2.7 ± 2.6 p=0.02 [4.6]**	1.2 [5.4]	0.9 ± 2.2 p=0.47 [3.6]
Eastern North America 5908 flights for 1994-2004 1346 flights for 2011-2016 8391 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**3.1 [6.1]**	**1.4 ± 0.4 p<0.01 [2.8]**	**4.7 [9.2]**	**2.1 ± 0.5 p<0.01 [4.2]**	**1.4 [2.8]**	**0.7 ± 0.6 p=0.03 [1.3]**
	Free troposphere (700-300 hPa)	**3.7 [6.8]**	**1.7 ± 0.4 p<0.01 [3.1]**	**3.8 [7.0]**	**1.7 ± 0.6 p<0.01 [3.2]**	**3.1 [5.7]**	**1.4 ± 0.6 p=0.01 [2.6]**
	Upper troposphere (400-300 hPa)	**5.2 [9.0]**	**2.4 ± 0.7 p<0.01 [4.1]**	**5.8 [9.9]**	**2.6 ± 0.7 p<0.01 [4.5]**	**4.6 [7.8]**	**2.1 ± 1.1 p=0.01 [3.5]**
	Middle troposphere (650 hPa)	**2.6 [5.0]**	**1.2 ± 0.5 p<0.01 [2.3]**	**3.2 [6.0]**	**1.5 ± 0.7 p<0.01 [2.8]**	**2.0 [3.8]**	**0.9 ± 0.7 p=0.01 [1.7]**
	Lower tropopshere (950-800 hPa)	**2.4 [5.7]**	**1.1 ± 0.7 p<0.01 [2.6]**	**6.0 [14.3]**	**2.7 ± 0.7 p<0.01 [6.5]**	**-2.0 [-4.8]**	**-0.9 ± 0.9 p=0.05 [-2.2]**
	OMI/MLS (TCO)	**2.6 [5.7]**	**1.7 ± 1.7 p=0.03 [3.8]**	**4.4 [6.5]**	**3.1 ± 1.9 p<0.01 [4.3]**	0.5 [5.1]	0.3 ± 2 p=0.73 [3.4]
Western North America 527 flights for 1994-2004 327 flights for 2011-2016 1569 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**3.5 [7.4]**	**1.6 ± 0.7 p<0.01 [3.4]**	**4.1 [8.6]**	**1.8 ± 0.9 p<0.01 [3.9]**	2.2 [4.7]	1.0 ± 2.2 p=0.39 [2.1]
	Free troposphere (700-300 hPa)	**6.2 [11.9]**	**2.8 ± 1.1 p<0.01 [5.4]**	**6.9 [13.3]**	**3.1 ± 1.1 p<0.01 [6.0]**	4.4 [8.6]	2.0 ± 2.9 p=0.20 [3.9]
	Upper troposphere (400-300 hPa)	**10.5 [19.0]**	**4.8 ± 1.6 p<0.01 [8.7]**	**9.9 [17.9]**	**4.5 ± 1.5 p<0.01 [8.1]**	**15.3 [27.8]**	**7.0 ± 4.0 p<0.01 [12.7]**
	Middle troposphere (650 hPa)	**2.9 [5.8]**	**1.3 ± 0.9 p=0.01 [2.7]**	**5.1 [10.4]**	**2.3 ± 0.8 p<0.01 [4.7]**	-4.7 [-9.6]	-2.1 ± 2.7 p=0.11 [-4.3]
	Lower tropopshere (950-800 hPa)	-0.4 [-1.1]	-0.2 ± 0.9 p=0.65 [-0.5]	1.2 [3.4]	0.6 ± 0.9 p=0.24 [1.6]	-5.3 [-14.7]	-2.4 ± 3.1 p=0.11 [-6.7]
	OMI/MLS (TCO)	**3.4 [6.6]**	**2.2 ± 1 p<0.01 [4.4]**	**3.9 [7.5]**	**2.6 ± 1.2 p<0.01 [5.0]**	**3.4 [5.9]**	**2.2 ± 1.5 p<0.01 [3.9]**
Northeast China / Korea 1440 flights for 1994-2004 920 flights for 2011-2016 2580 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**7.2 [12.8]**	**3.3 ± 0.7 p<0.01 [5.8]**	**4.7 [8.3]**	**2.1 ± 0.7 p<0.01 [3.8]**	**11.7 [20.8]**	**5.3 ± 1.6 p<0.01 [9.5]**
	Free troposphere (700-300 hPa)	**3.8 [6.6]**	**1.7 ± 0.7 p<0.01 [3.0]**	**2.3 [3.9]**	**1.0 ± 0.5 p<0.01 [1.8]**	**7.2 [12.4]**	**3.3 ± 1.3 p<0.01 [5.7]**
	Upper troposphere (400-300 hPa)	**4.1 [6.5]**	**1.8 ± 0.9 p<0.01 [2.9]**	1.3 [2.0]	0.6 ± 1.2 p=0.35 [0.9]	**7.5 [12.0]**	**3.4 ± 1.8 p<0.01 [5.5]**
	Middle troposphere (650 hPa)	**5.1 [9.3]**	**2.3 ± 0.6 p<0.01 [4.2]**	**3.6 [6.6]**	**1.7 ± 0.6 p<0.01 [3.0]**	**6.7 [12.2]**	**3.1 ± 1.1 p<0.01 [5.6]**
	Lower tropopshere (950-800 hPa)	**13.8 [27.3]**	**6.3 ± 1.5 p<0.01 [12.4]**	**8.0 [15.7]**	**3.6 ± 1.2 p<0.01 [7.2]**	**22.1 [43.7]**	**10.0 ± 2.3 p<0.01 [19.9]**
	OMI/MLS (TCO)	**4.8 [5.7]**	**3.2 ± 1.4 p<0.01 [3.8]**	**5.6 [6.5]**	**3.8 ± 1 p<0.01 [4.3]**	**5.1 [5.0]**	**3.4 ± 1.4 p<0.01 [3.4]**
Southeast US 2402 flights for 1994-2004 342 flights for 2011-2016 3528 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	*1.4 [2.8]*	*0.6 ± 0.8 p=0.09 [1.3]*	**2.6 [5.0]**	**1.2 ± 1.0 p=0.02 [2.3]**	-0.1 [-0.1]	-0.0 ± 1.4 p=0.97 [-0.0]
	Free troposphere (700-300 hPa)	**2.9 [5.6]**	**1.3 ± 0.9 p<0.01 [2.5]**	*3.0 [5.8]*	*1.4 ± 1.4 p=0.05 [2.7]*	**2.9 [5.5]**	**1.3 ± 1.1 p=0.04 [2.5]**
	Upper troposphere (400-300 hPa)	1.4 [2.6]	0.7 ± 1.5 p=0.39 [1.2]	2.2 [4.0]	1.0 ± 1.6 p=0.20 [1.8]	1.0 [1.9]	0.5 ± 2.4 p=0.70 [0.9]
	Middle troposphere (650 hPa)	1.2 [2.3]	0.5 ± 0.7 p=0.14 [1.0]	0.7 [1.4]	0.3 ± 1.1 p=0.55 [0.6]	1.7 [3.4]	0.8 ± 1.1 p=0.14 [1.6]
	Lower tropopshere (950-800 hPa)	0.4 [0.8]	0.2 ± 1.2 p=0.82 [0.4]	**2.5 [5.4]**	**1.2 ± 1.1 p=0.05 [2.4]**	-1.7 [-3.6]	-0.8 ± 2.6 p=0.56 [-1.6]
	OMI/MLS (TCO)	**4.6 [5.6]**	**2.9 ± 2.4 p=0.01 [3.7]**	**5.8 [6.8]**	**3.9 ± 2.4 p<0.01 [4.5]**	0.8 [4.7]	0.7 ± 1.7 p=0.46 [3.2]
Persian Gulf 1247 flights for 1998-2005 1077 flights for 2011-2016 2345 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**7.0 [12.6]**	**3.2 ± 1.0 p<0.01 [5.7]**	*2.0 [3.5]*	*0.9 ± 0.9 p=0.05 [1.6]*	**12.3 [22.2]**	**5.6 ± 1.1 p<0.01 [10.1]**
	Free troposphere (700-300 hPa)	**6.6 [11.6]**	**3.0 ± 0.9 p<0.01 [5.3]**	-0.7 [-1.1]	-0.3 ± 1.4 p=0.66 [-0.5]	**12.4 [21.7]**	**5.6 ± 1.5 p<0.01 [9.9]**
	Upper troposphere (400-300 hPa)	**6.3 [10.3]**	**2.9 ± 1.4 p<0.01 [4.7]**	-0.8 [-1.3]	-0.3 ± 2.4 p=0.77 [-0.6]	**11.3 [18.5]**	**5.2 ± 1.8 p<0.01 [8.4]**
	Middle troposphere (650 hPa)	**8.6 [16.3]**	**3.9± 1.5 p<0.01 [7.4]**	1.5 [2.8]	0.7 ± 1.2 p=0.25 [1.3]	**15.2 [28.5]**	**6.9 ± 1.9 p<0.01 [13.0]**
	Lower tropopshere (950-800 hPa)	**10.8 [21.1]**	**4.9 ± 0.9 p<0.01 [9.6]**	**6.5 [12.7]**	**3.0 ± 1.3 p<0.01 [5.8]**	**15.6 [30.5]**	**7.1 ± 1.8 p<0.01 [13.9]**
	OMI/MLS (TCO)	**3.7 [6.0]**	**2.4 ± 1.1 p<0.01 [4.0]**	**2.9 [5.9]**	**2.7 ± 1.4 p=0.01 [3.9]**	**4 [6.0]**	**2.7 ± 1.6 p<0.01 [4.0]**

Table S1b. Following Table S1a with the five tropical regions. *Due to the variability of the terrain across northern South America, the tropospheric column is limited to 600-250 hPa above this region.*

	Pressure level	Annual		Cold season		Warm Season	
		nmol mol^{-1} [%]	**nmol mol^{-1} decade^{-1} [% decade^{-1}]**	**nmol mol^{-1} [%]**	**nmol mol^{-1} decade^{-1} [% decade^{-1}]**	**nmol mol^{-1} [%]**	**nmol mol^{-1} decade^{-1} [% decade^{-1}]**
Northern South America 874 flights for 1994-2004 349 flights for 2011-2016 1627 flights for 1994-2016	**Tropospheric column (600-250 hPa)**	**3.1 [8.0]**	**1.4 ± 1.0 p=0.01 [3.6]**	**5.7 [14.8]**	**2.6 ± 1.0 p<0.01 [6.7]**	-1.9 [-4.8]	-0.9 ± 2.0 p=0.42 [-2.2]
	Free troposphere (600-300 hPa)	**3.2 [8.4]**	**1.5 ± 1.2 p=0.01 [3.8]**	**5.6 [14.5]**	**2.5 ± 1.0 p<0.01 [6.6]**	-2.3 [-5.9]	-1.0 ± 1.8 p=0.26 [-2.7]
	Upper troposphere (400-300 hPa)	**3.6 [8.8]**	**1.6 ± 1.0 p<0.01 [4.0]**	**6.9 [17.2]**	**3.2 ± 1.3 p<0.01 [7.8]**	-0.5 [-1.3]	-0.3 ± 1.8 p= 0.78 [-0.6]
	Middle troposphere (650 hPa)	NA	NA	NA	NA	NA	NA
	Lower tropopshere (950-800 hPa)	NA	NA	NA	NA	NA	NA
	OMI/MLS (TCO)	**5 [5.6]**	**3.3 ± 1 p<0.01 [3.7]**	**3.6 [6.3]**	**2.3 ± 1.3 p<0.01 [4.2]**	**5.8 [5.0]**	**3.7 ± 0.7 p<0.01 [3.3]**
Gulf of Guinea 928 flights for 1994-2004 480 flights for 2011-2016 1427 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**4.7 [10.4]**	**2.1 ± 1.1 p<0.01 [4.7]**	**5.6 [12.4]**	**2.5 ± 1.5 p<0.01 [5.6]**	*3.9 [8.7]*	*1.8 ± 2.0 p=0.05 [4.0]*
	Free troposphere (700-300 hPa)	*2.7 [5.3]*	*1.2 ± 1.4 p=0.07 [2.4]*	1.7 [3.3]	0.8 ± 1.5 p=0.35 [1.5]	4.3 [8.5]	1.9 ± 2.3 p=0.12 [3.9]
	Upper troposphere (400-300 hPa)	0.5 [1.0]	0.2 ± 1.4 p=0.72 [0.5]	1.4 [2.5]	0.6 ± 2.0 p=0.54 [1.2]	-0.6 [-1.0]	-0.3 ± 3.1 p=0.86 [-0.5]
	Middle troposphere (650 hPa)	**5.8 [12.2]**	**2.7 ± 1.6 p<0.01 [5.6]**	3.2 [6.8]	1.5 ± 1.9 p=0.18 [3.1]	**8.5 [17.7]**	**3.9 ± 2.1 p<0.01 [8.1]**
	Lower tropopshere (950-800 hPa)	**12.7 [38.9]**	**5.8 ± 1.8 p<0.01 [17.7]**	**16.0 [49.2]**	**7.3 ± 2.3 p<0.01 [22.4]**	**9.7 [29.6]**	**4.4 ± 1.8 p<0.01 [13.5]**
	OMI/MLS (TCO)	**4.3 [7.6]**	**2.9 ± 0.7 p<0.01 [5.0]**	**4 [7.7]**	**2.6 ± 1 p<0.01 [5.1]**	**4.3 [7.5]**	**2.9 ± 1.3 p<0.01 [5.0]**
India 434 flights for 1994-2004 389 flights for 2011-2016 1351 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**10.4 [24.3]**	**4.7 ± 1.2 p<0.01 [11.0]**	**11.3 [26.3]**	**5.1 ± 2.1 p<0.01 [12.0]**	**10.1 [23.6]**	**4.6 ± 1.6 p<0.01 [10.7]**
	Free troposphere (700-300 hPa)	**8.0 [16.8]**	**3.6 ± 1.5 p<0.01 [7.6]**	**7.0 [14.8]**	**3.2 ± 2.0 p<0.01 [6.7]**	**8.5 [17.9]**	**3.9 ± 2.0 p<0.01 [8.1]**
	Upper troposphere (400-300 hPa)	**10.7 [21.0]**	**4.9 ± 2.2 p<0.01 [9.6]**	**7.2 [14.1]**	**3.3 ± 3.1 p=0.05 [6.4]**	**13.9 [27.3]**	**6.3 ± 3.2 p<0.01 [12.4]**
	Middle troposphere (650 hPa)	**7.1 [16.0]**	**3.2 ± 2.7 p=0.01 [7.3]**	*5.9 [13.1]*	*2.7 ± 3.0 p=0.08 [6.0]*	**9.3 [20.9]**	**4.4 ± 3.7 p=0.01 [9.5]**
	Lower tropopshere (950-800 hPa)	**18.4 [57.5]**	**8.3 ± 1.5 p<0.01 [26.2]**	**23.7 [74.0]**	**10.8 ± 2.3 p<0.01 [33.6]**	**16.2 [50.8]**	**7.4 ± 1.8 p<0.01 [23.1]**
	OMI/MLS (TCO)	**7 [6.2]**	**4.7 ± 1.6 p<0.01 [4.1]**	**6.2 [6.1]**	**3.9 ± 1.9 p<0.01 [4.1]**	**6.8 [6.3]**	**4.5 ± 2 p<0.01 [4.2]**
Southeast Asia 952 flights for 1994-2004 1374 flights for 2011-2016 2744 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**11.9 [29.7]**	**5.4 ± 1.2 p<0.01 [13.5]**	**7.4 [19.3]**	**3.5 ± 1.1 p<0.01 [8.8]**	**20.3 [50.5]**	**9.2 ± 1.3 p<0.01 [22.9]**
	Free troposphere (700-300 hPa)	**12.3 [29.6]**	**5.6 ± 1.0 p<0.01 [13.5]**	**7.0 [16.9]**	**3.2 ± 0.9 p<0.01 [7.7]**	**20.9 [50.5]**	**9.2 ± 1.3 p<0.01 [23.3]**
	Upper troposphere (400-300 hPa)	**13.4 [28.6]**	**6.1 ± 1.2 p<0.01 [13.0]**	**8.2 [17.5]**	**3.7 ± 1.4 p<0.01 [8.0]**	**21.2 [45.5]**	**9.6 ± 2.2 p<0.01 [20.7]**
	Middle troposphere (650 hPa)	**13.8 [36.8]**	**6.3 ± 1.0 p<0.01 [16.7]**	**6.4 [17.2]**	**2.9 ± 1.4 p<0.01 [7.8]**	**20.1 [53.7]**	**9.2 ± 1.5 p<0.01 [24.4]**
	Lower tropopshere (950-800 hPa)	**12.0 [32.5]**	**5.4 ± 0.9 p<0.01 [14.8]**	**7.3 [19.7]**	**3.3 ± 2.0 p<0.01 [9.0]**	**16.7 [45.2]**	**7.6 ± 1.5 p<0.01 [20.6]**
	OMI/MLS (TCO)	**7 [6.7]**	**4.7 ± 2.3 p<0.01 [4.4]**	**4.9 [6.6]**	**3.3 ± 2.6 p=0.02 [4.4]**	**7.5 [6.5]**	**5 ± 2.3 p<0.01 [4.3]**
Malaysia / Indonesia 169 flights for 1995-2000 368 flights for 2012-2016 581 flights for 1994-2016	**Tropospheric column (950-250 hPa)**	**12.7 [40.8]**	**5.8 ± 1.7 p<0.01 [18.5]**	**14.6 [47.1]**	**6.6 ± 1.5 p<0.01 [21.4]**	**7.3 [23.6]**	**3.3 ± 2.3 p=0.01 [10.7]**
	Free troposphere (700-300 hPa)	**7.6 [24.6]**	**3.5 ± 1.3 p<0.01 [11.2]**	**9.2 [29.7]**	**4.2 ± 1.6 p<0.01 [13.5]**	6.4 [20.7]	2.9 ± 3.0 p=0.05 [9.4]
	Upper troposphere (400-300 hPa)	**6.2 [17.9]**	**2.8 ± 2.2 p=0.01 [8.1]**	5.2 [15.3]	2.4 ± 3.3 p=0.16 [6.9]	*5.7 [16.5]*	*2.6 ± 3.3 p=0.08 [7.5]*
	Middle troposphere (650 hPa)	**10.8 [38.4]**	**4.9 ± 1.7 p<0.01 [17.5]**	**14.8 [52.5]**	**6.7 ± 1.7 p<0.01 [23.9]**	**6.2 [21.9]**	**2.8 ± 2.3 p=0.03 [10.0]**
	Lower tropopshere (950-800 hPa)	**17.5 [57.9]**	**8.0 ± 2.5 p<0.01 [26.3]**	**21.5 [71.1]**	**9.8 ± 3.2 p<0.01 [32.3]**	**13.0 [43.0]**	**5.9 ± 4.1 p=0.01 [19.5]**
	OMI/MLS (TCO)	**4.2 [8.8]**	**2.9 ± 1.7 p<0.01 [5.8]**	**8.2 [8.7]**	**5.5 ± 3.6 p=0.01 [5.8]**	**4.2 [8.8]**	**2.7 ± 1.7 p<0.01 [5.9]**

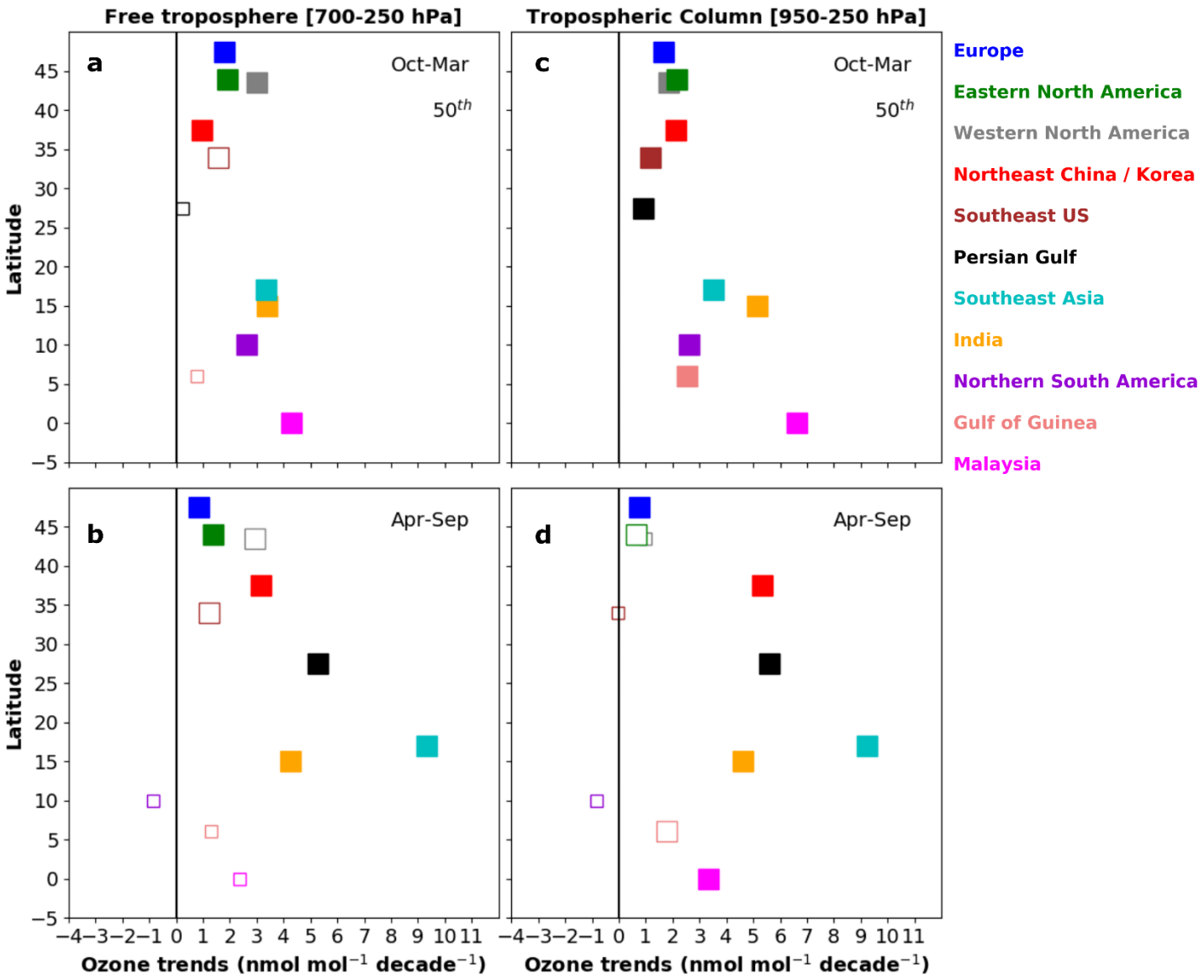

Figure S1. Seasonal trends of median ozone (nmol mol^{-1} decade^{-1}) for the free troposphere (700-300 hPa, panel a,b) and the tropospheric column (950-250 hPa, panel c,d). *The trends are calculated between 1994 and 2016 above Western North America (grey), Eastern North America (green), Europe (blue), Northeast China / Korea (red), Southeast US (brown), Northern South America (purple), Gulf of Guinea (salmon), India (orange), Southeast Asia (cyan), between 1998 and 2016 above the Persian Gulf (black), and between 1995 and 2016 above Malaysia (magenta). Due to the variability of the terrain across northern South America, the tropospheric column is limited to 600-250 hPa above this region. Large squares indicate trends with p-values less than 0.05, large open squares indicate trends with p-values between 0.05 and 0.1 and small open squares indicate p-values greater than 0.1.*

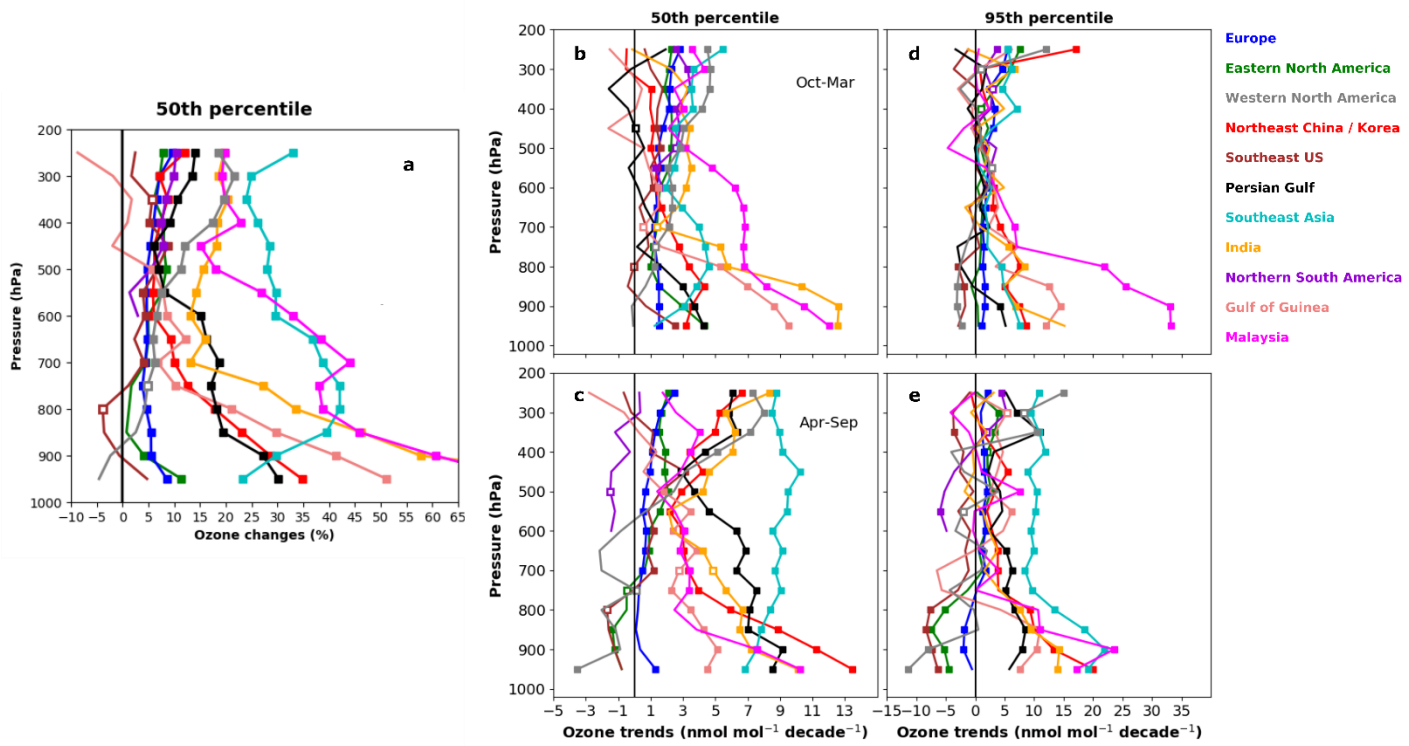

Figure S2. Annual trends of the 50th percentile of ozone in percent over the entire time-period 1994-2016 and seasonal trends of the 50th and 95th percentiles of ozone (nmol mol^{-1} decade^{-1}) at intervals of 50 hPa. *The trends are calculated on each pressure level (every 50 hPa) between 1994 and 2016 above Western North America (grey), Eastern North America (green), Europe (blue), Northeast China / Korea (red), Southeast US (brown), India (orange), Southeast Asia (cyan), Northern South America (purple), Gulf of Guinea (salmon), between 1998 and 2016 above the Persian Gulf (black), and between 1995 and 2016 above Malaysia/Indonesia (magenta), annually (a), for the cold season October-March (b,d) and the warm season April-September (c,e). Squares indicate trends with p-values less than 0.05. Open squares indicate trends with p-values between 0.05 and 0.1.*

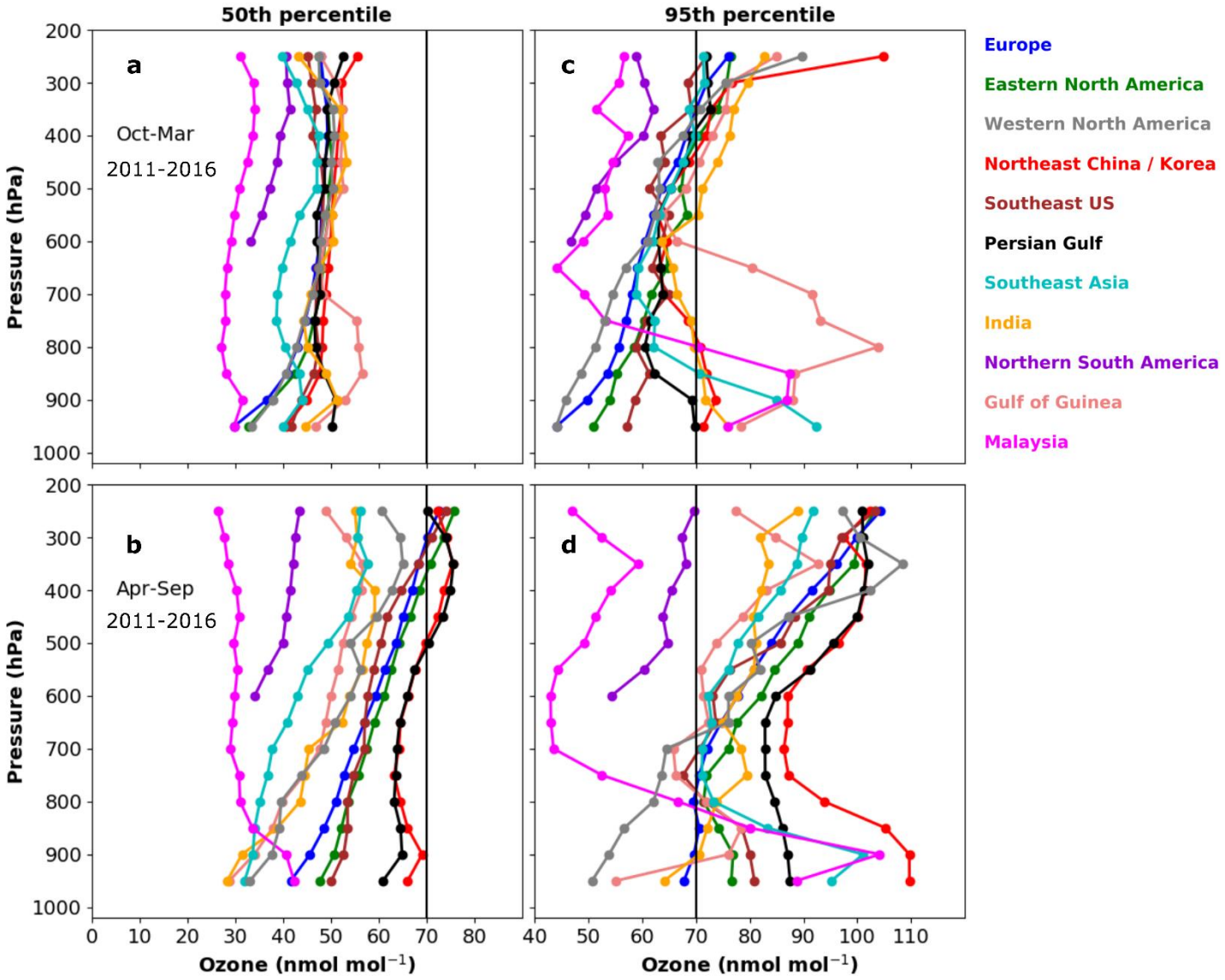

Figure S3. Seasonal vertical profiles of the 50th (panels a,b) and 95th percentiles (panels c,d) of ozone (nmol mol^{-1}). *The vertical profiles of ozone are measured by IAGOS commercial aircraft above Western North America (grey), Eastern North America (green), Europe (blue), Northeast China / Korea (red), Southeast US (brown), Northern South America (purple), Gulf of Guinea (Salmon), the Persian Gulf (black), India (orange), Southeast Asia (cyan) and Malaysia/Indonesia (magenta) between 2011 and 2016, for the cold season October-March (a,c) and the warm season April-September (b,d). For reference, the 70 nmol mol^{-1} value is indicated with a vertical black line, which corresponds to the United States National Ambient Air Quality Standards for ozone.*

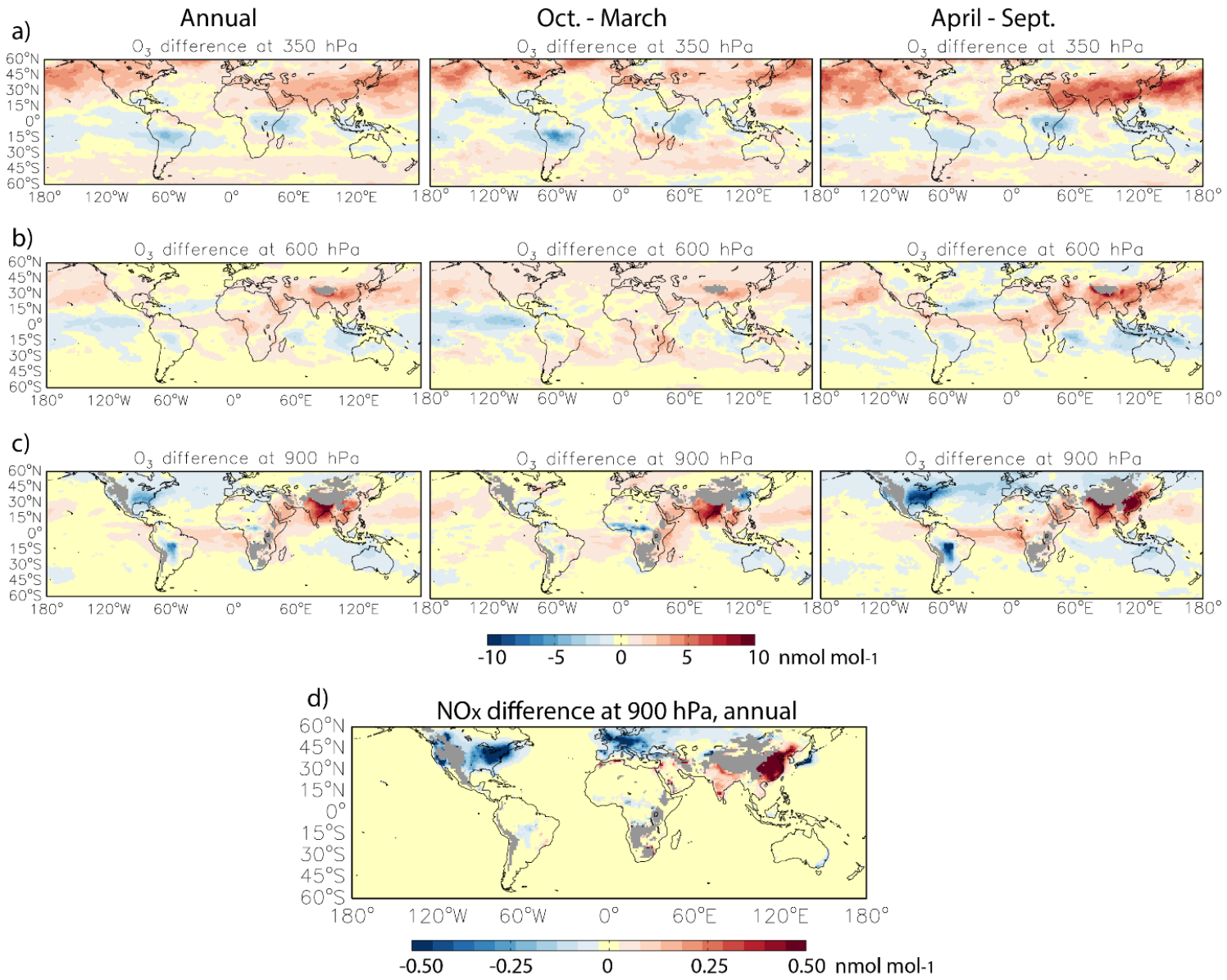

Figure S4. Differences of ozone and NO_x between 1994-2004 and 2011-2016 based on annual and seasonal (October-March and April-September) data as simulated by the MERRA-2 GMI global atmospheric chemistry model. *The differences of ozone are calculated on three pressure surfaces: a) 350 hPa, b) 600 hPa and c) 900 hPa. The difference for NOx is only shown at 900 hPa (d).*

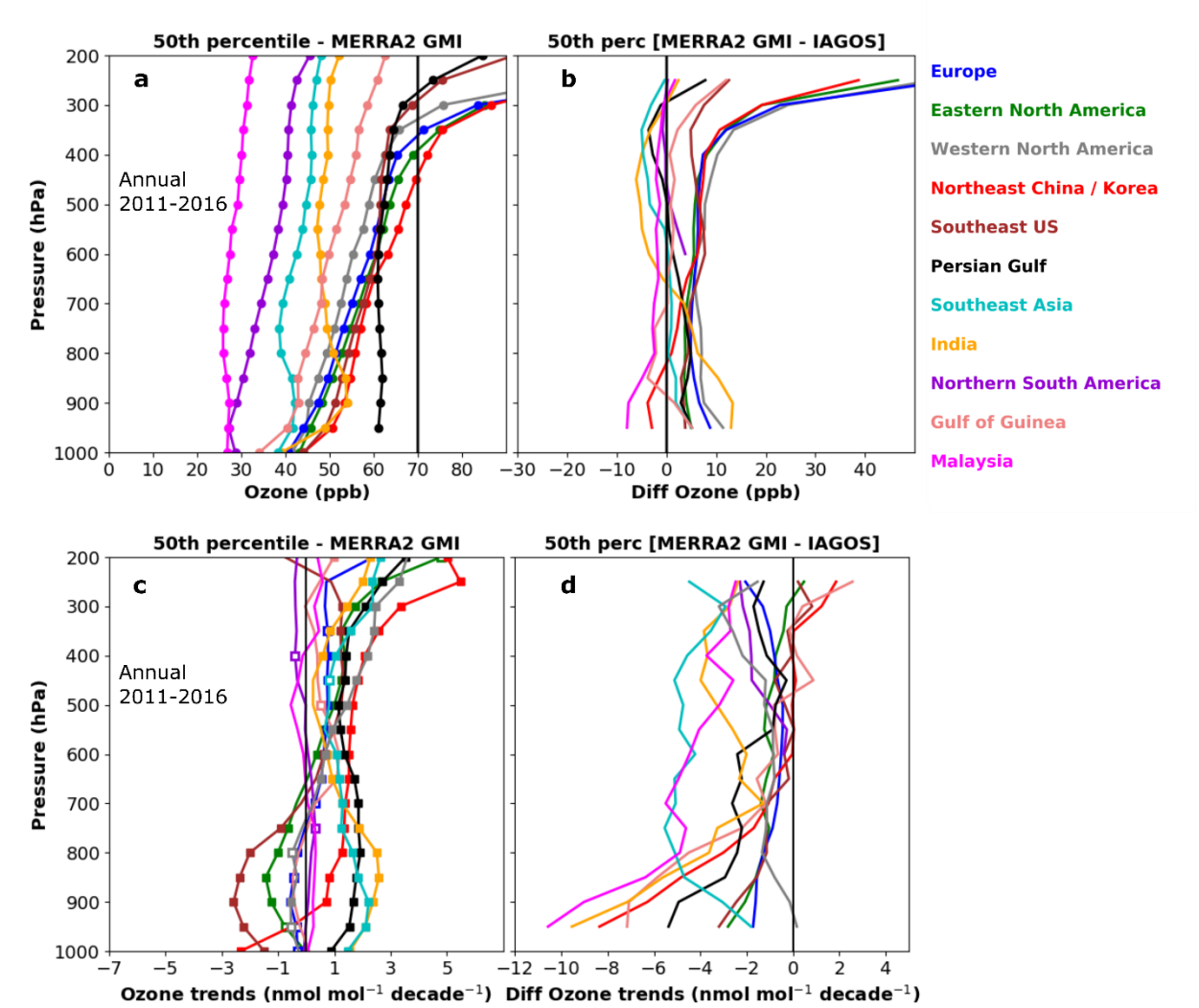

Figure S5. ***Annual distributions (nmol mol^{-1}) and trends (nmol mol^{-1} decade^{-1}) of the 50th percentiles at intervals of 50 hPa from the MERRA2 GMI model, and the differences in the profiles between the MERRA2 GMI model and the IAGOS observations.*** *The profiles of ozone (a, b) are shown for the recent period 2011-2016 above the eleven regions. For reference, the 70 nmol mol^{-1} value is indicated with a vertical black line, which corresponds to the United States National Ambient Air Quality Standards for ozone. The trends (c, d) are calculated between 1994 and 2016 above Western North America (grey), Eastern North America (green), Europe (blue), Northeast China / Korea (red), Southeast US (brown), Northern South America (purple), Gulf of Guinea (Salmon), India (orange), Southeast Asia (cyan), between 1998 and 2016 above the Persian Gulf (black), and between 1995 and 2016 above Malaysia/Indonesia (magenta). Squares indicate trends with p-values less than 0.05. Open squares indicate trends with p-values between 0.05 and 0.1. The zero trend value is indicated with a vertical black line. For the differences in the trends (panels to the right) between the model and the observations, the black line marks the zero value indicating no differences in trends between the two datasets.*

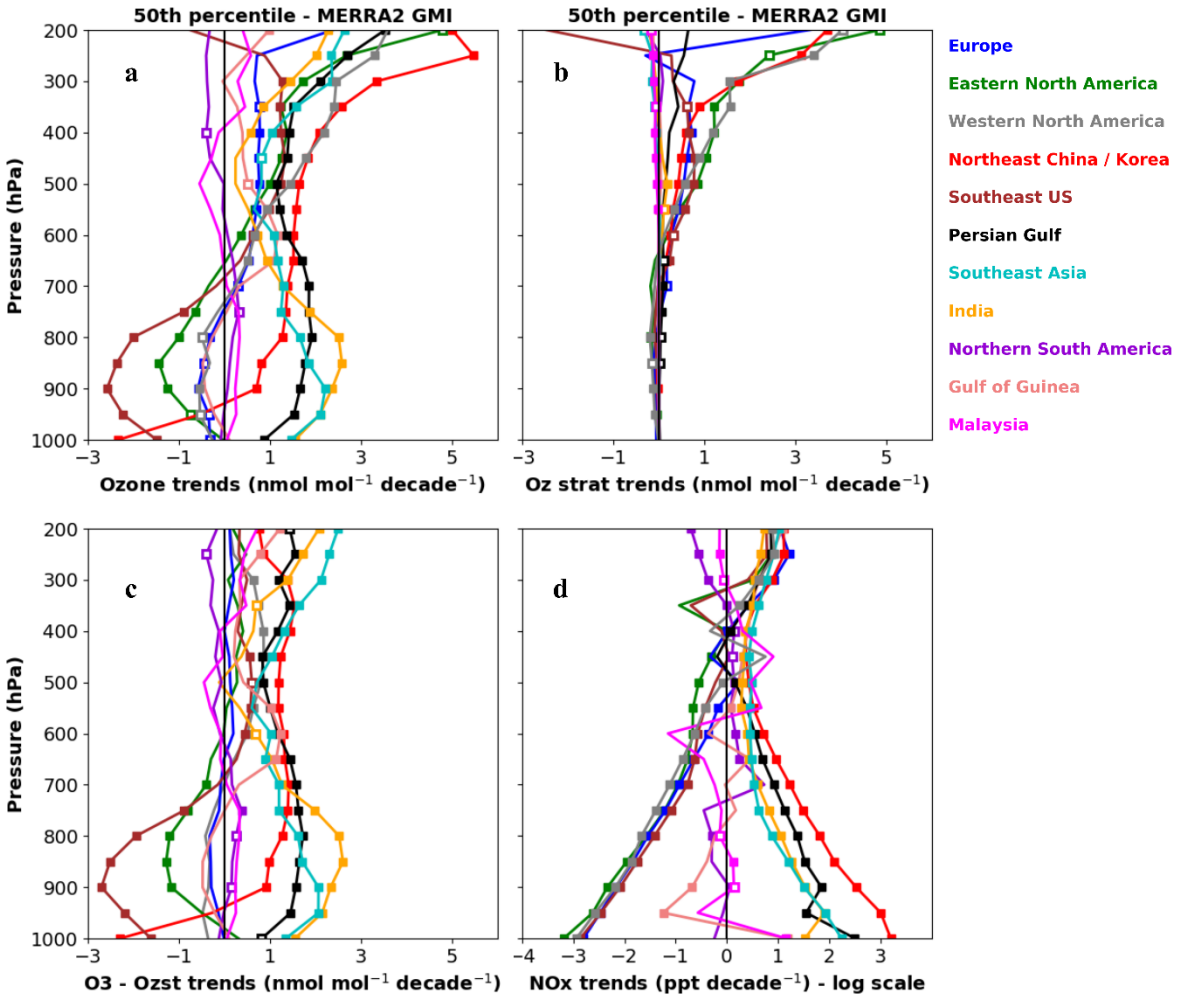

Figure S6. Annual trends (nmol mol^{-1} decade^{-1}) of tropospheric ozone (a), stratospheric ozone tracer (b), tropospheric ozone minus stratospheric ozone tracer for the 50th percentile (c), and NOx trends (ppt decade^{-1}) from MERRA2-GMI (d). *The trends are calculated between 1994 and 2016 above Western North America (grey), Eastern North America (green), Europe (blue), Northeast China / Korea (red), Southeast US (brown), Northern South America (purple), Gulf of Guinea (Salmon), India (orange), Southeast Asia (cyan), between 1998 and 2016 above the Persian Gulf (black), and between 1995 and 2016 above Malaysia/Indonesia (magenta). Squares indicate trends with p-values less than 0.05. Open squares indicate trends with p-values between 0.05 and 0.1. The zero trend value is indicated with a vertical black line. Note the log scale for the trends of NOx.*

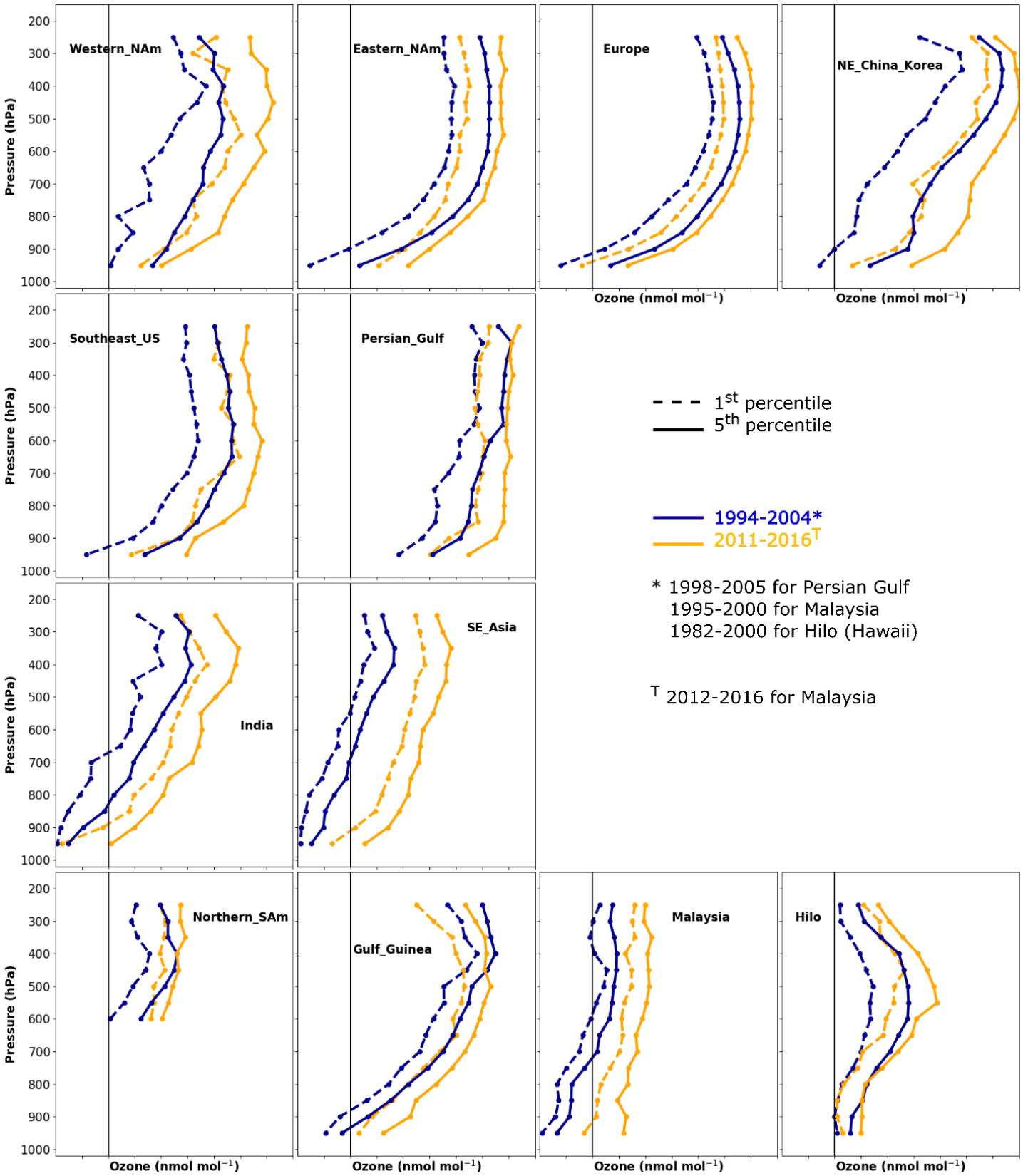

Figure S7. Vertical profiles of the 1st and the 5th percentiles of ozone*. The periods 1994-2004 (dark blue) and 2011-2016 (orange) are shown above Western North America, Eastern North America, Europe, Northeast China / Korea, Southeast US, Persian Gulf, India, Southeast Asia, Northern South America, Gulf of Guinea,*

Malaysia/Indonesia and Hilo (Hawaï). The earlier period is limited to 1998-2005 for Persian Gulf and 1995-2000 for Malaysia/Indonesia due to data availability, and is extended to 1982-2000 for Hilo (Hawaii). The vertical black bar indicates the 10 nmol mol^{-1} ozone value.

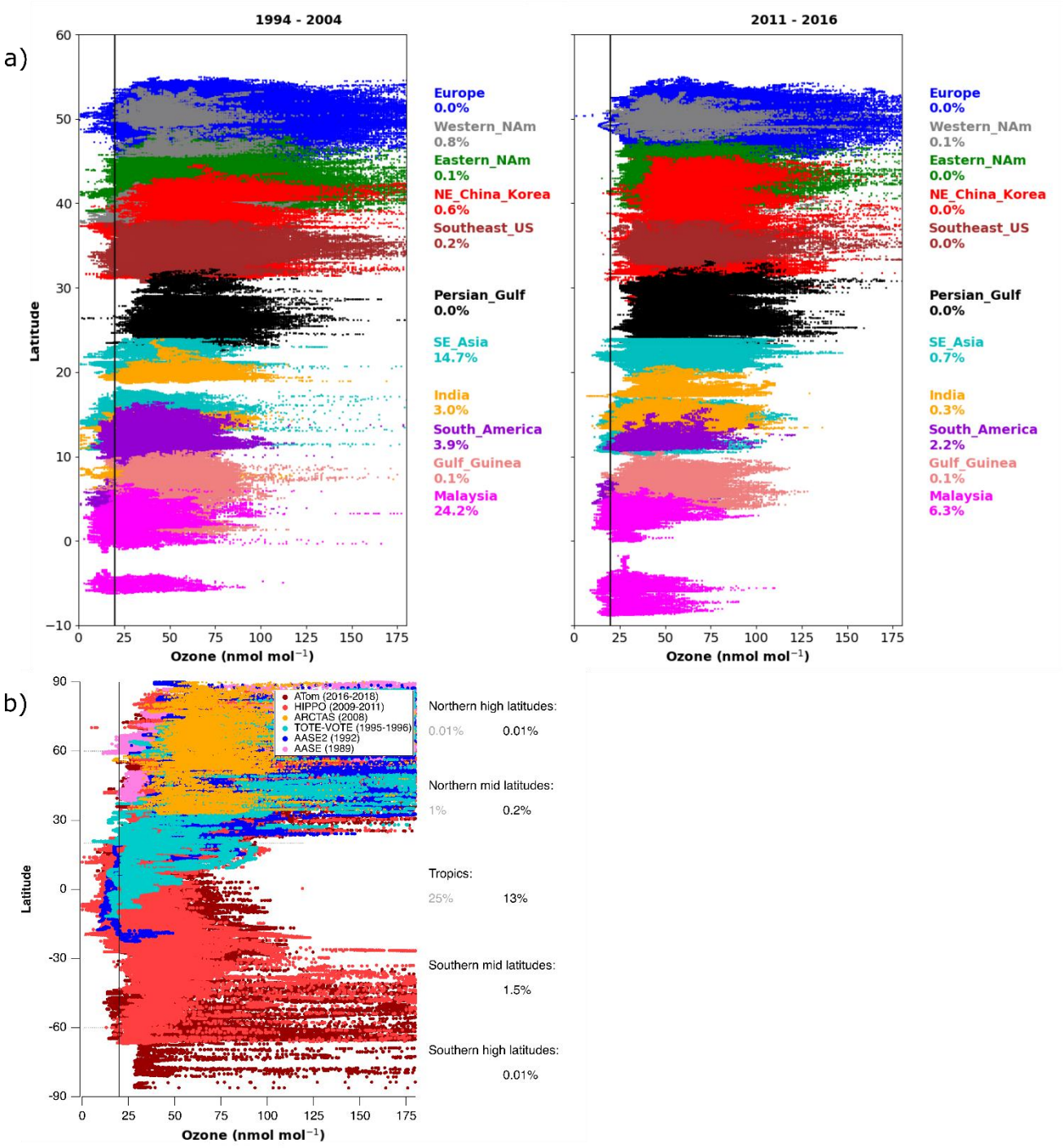

Figure S8 a) A comparison of IAGOS free tropospheric ozone observations (above the 700 hPa level) between 1994-2004 (left) and 2011-2016 (right) and b) Free tropospheric ozone observations between 700 hPa and 100 hPa from six aircraft campaigns conducted above the North and South Pacific Oceans between 1989 and 2018. *Also shown are the percentages of ozone values in each region below 20 nmol mol^{-1}: Western North America (grey), Eastern North America (green), Europe (blue), Northeast China / Korea (red), Southeast US (brown), India (orange), Southeast Asia (cyan), South America (purple), Gulf of Guinea (salmon), Persian Gulf (black), and above Malaysia/Indonesia (magenta), and per latitude band for 1989-1996 (grey) and 2008-2018 (black): Northern high latitudes (60-90°N), Northern mid-latitudes (20-60°N), Tropics (20°S-20°N), Southern mid-latitudes (20-60°S) and Southern high latitudes (60-90°S). The vertical black line indicates the 20 nmol mol^{-1} value.*

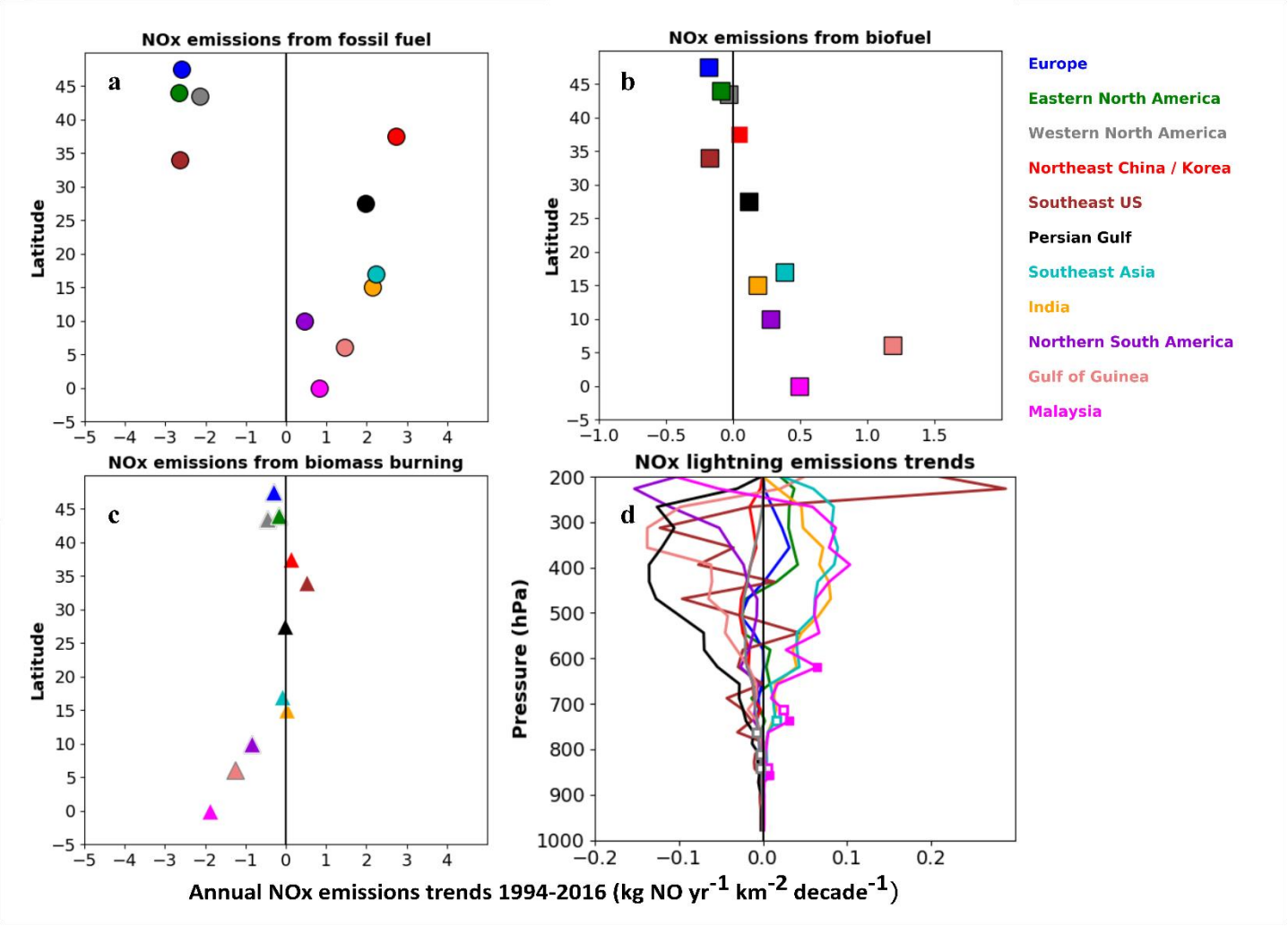

Figure S9. Annual NOx emissions trends from fossil fuel, biofuel, biomass burning and lightning (kg NO yr^{-1} km^{-2} $decade^{-1}$). *These emissions inventories are used in the MERRA-2 GMI global model simulation. The trends are calculated for the time period 1994-2016 and for the eleven regions, Western North America (grey), Eastern North America (green), Europe (blue), Northeast China / Korea (red), Southeast US (brown), Northern South America (purple), Gulf of Guinea (Salmon), the Persian Gulf (black), India (orange), Southeast Asia (cyan) and Malaysia (magenta). Note the different scale for biofuel and lightning emissions.*