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Supplement of

Long-term change in the source contribution to surface ozone over Japan

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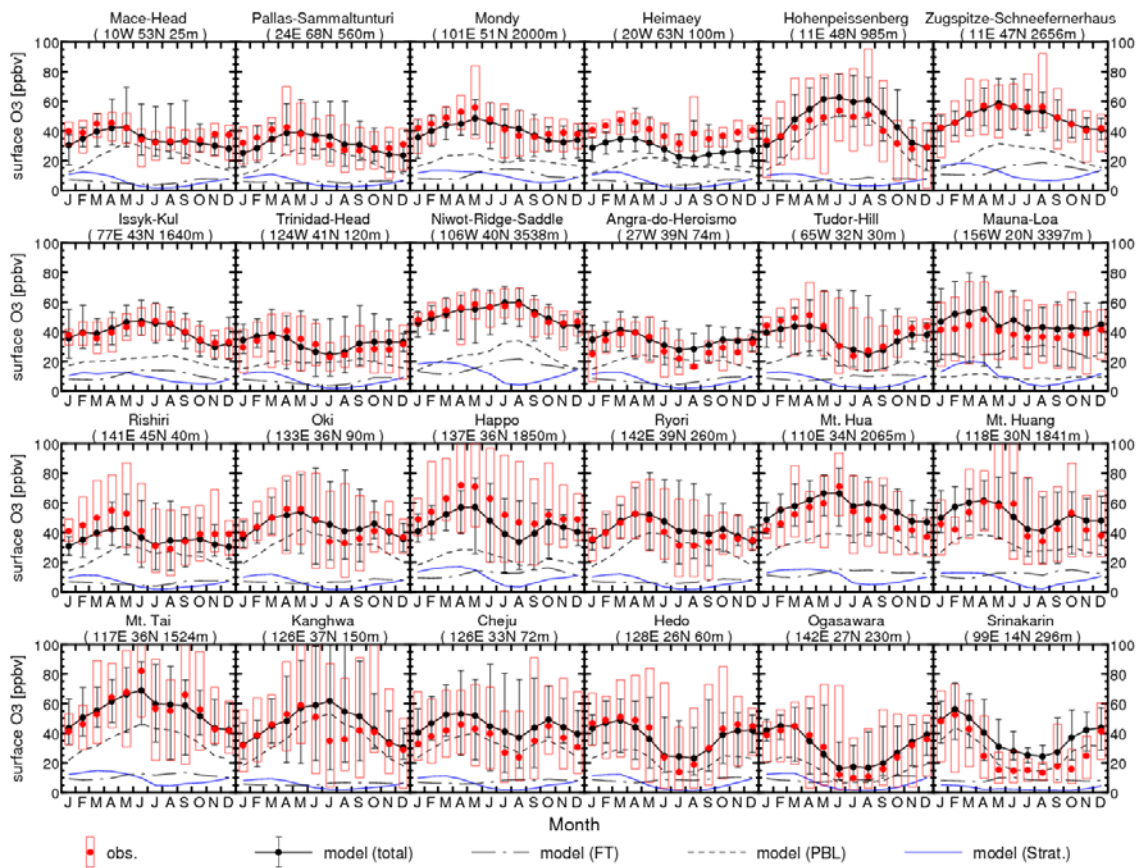


Figure S1. Seasonal variations of the monthly mean surface O_3 observations (red filled circles) and model calculations (black circles and thick black solid lines), with contributions from the stratosphere (blue solid lines), PBL (black dashed lines), and FT (black dash-dotted lines) estimated by the model. Monthly means and each contribution are multiyear (six years for the model and available years for the observations) averaged values for 2000–2005. Vertical bars and boxes denote the multiyear range of the daily mean surface O_3 for each month for the model and the observations, respectively.

Table S1. Statistical summary of the comparison between the observed (Obs) and modeled (Model) surface O₃ at observational sites shown in Figure S1. R is correlation coefficients, MB is mean bias, and RMSE is root mean square error.

Station name	Mean (Obs) [ppbv]	Mean (Model) [ppbv]	R	MB [ppbv]	RMSE [ppbv]
Mace-Head	37.81	34.5	0.63	-3.31	5.08
Pallas-Sammaltunturi	33.02	31.38	0.6	-1.64	4.95
Mondy	43.5	40.08	0.8	-3.42	5.13
Heimaey	39.75	28.12	0.87	-11.63	11.83
Hohenpeissenberg	41.05	47.48	0.94	6.43	8.27
Zugspitze-Schneefernerhaus	49.81	49.33	0.97	-0.48	1.5
Issyk-Kul	39.54	39.65	0.93	0.11	2.12
Trinidad-Head	30.92	32.09	0.67	1.17	3.95
Niwot-Ridge-Saddle	52.76	52.03	0.92	-0.73	2.29
Angra-do-Heroismo	29.49	34.26	0.88	4.77	6.05
Tudor-Hill	39.54	35.95	0.96	-3.59	4.74
Mauna-Loa	40.15	46.26	0.86	6.11	6.57
Rishiri	41.33	35.52	0.76	-5.81	7.97
Oki	43	44.51	0.8	1.51	4.94
Happo	55.58	45.43	0.92	-10.15	10.78
Ryori	39.06	42.45	0.83	3.39	5.03
Mt. Hua	50.29	56.69	0.92	6.4	7.59
Mt. Huang	47.23	51.47	0.77	4.24	7.1
Mt. Tai	56.18	55.16	0.93	-1.02	5.13
Kanghwa	41.33	46.29	0.58	4.96	10.24
Cheju	36.75	45.46	0.92	8.71	9.15
Hedo	38.42	37.01	0.96	-1.41	4.76
Ogasawara	28.08	30.28	0.96	2.2	4.15
Srinakaran	27.35	38	0.92	10.66	12.26

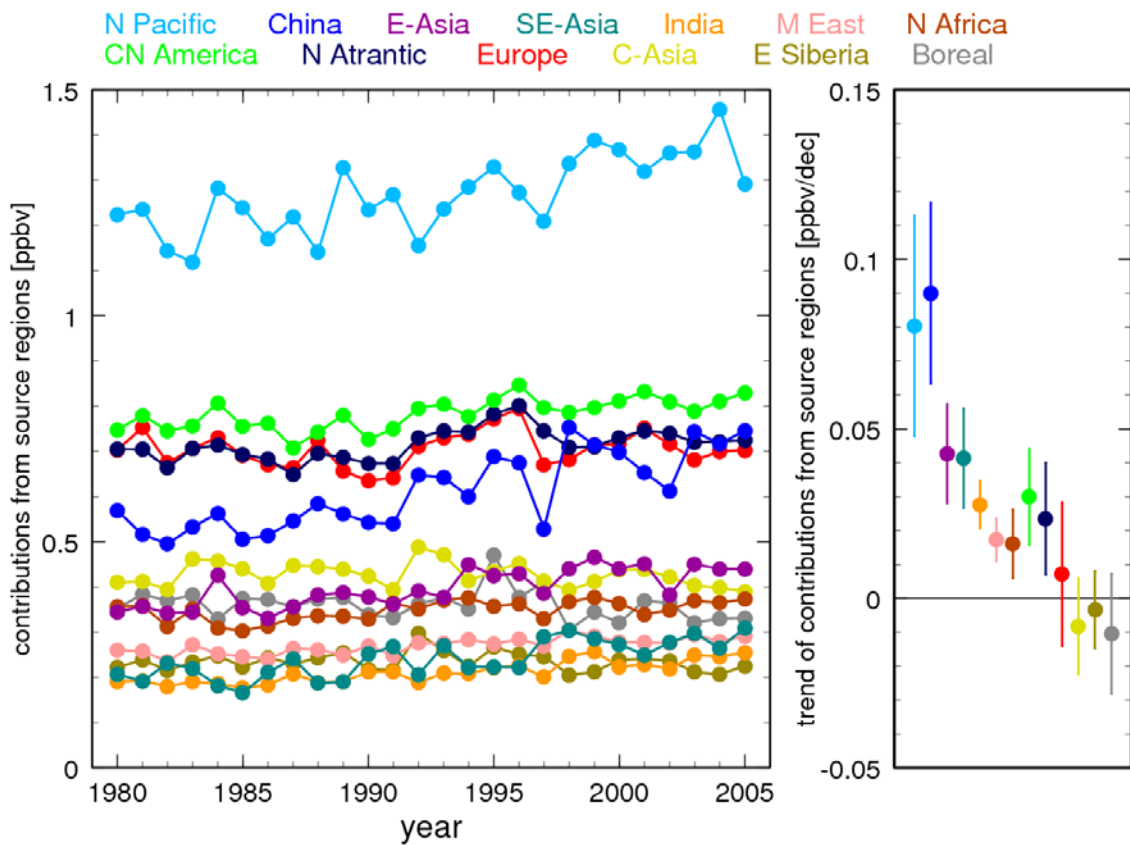


Figure S2. (Left) The long-term changes of annual mean contributions from source regions in the free troposphere in the Northern Hemisphere to surface O₃ over Japan. Some source regions are grouped: E-Asia is the sum of JPN, KOR, JPS, and ECS; SE-Asia is the sum of IDN and IDC; and CN America is the sum of AMC and AMN. (Right) The linear trends of the regional contributions in the left panel are for 1980–2005. Error bars are the 95 % confidence intervals.