Supplementary Material for

Deconstruction of tropospheric chemical reactivity using aircraft measurements: the ATom data

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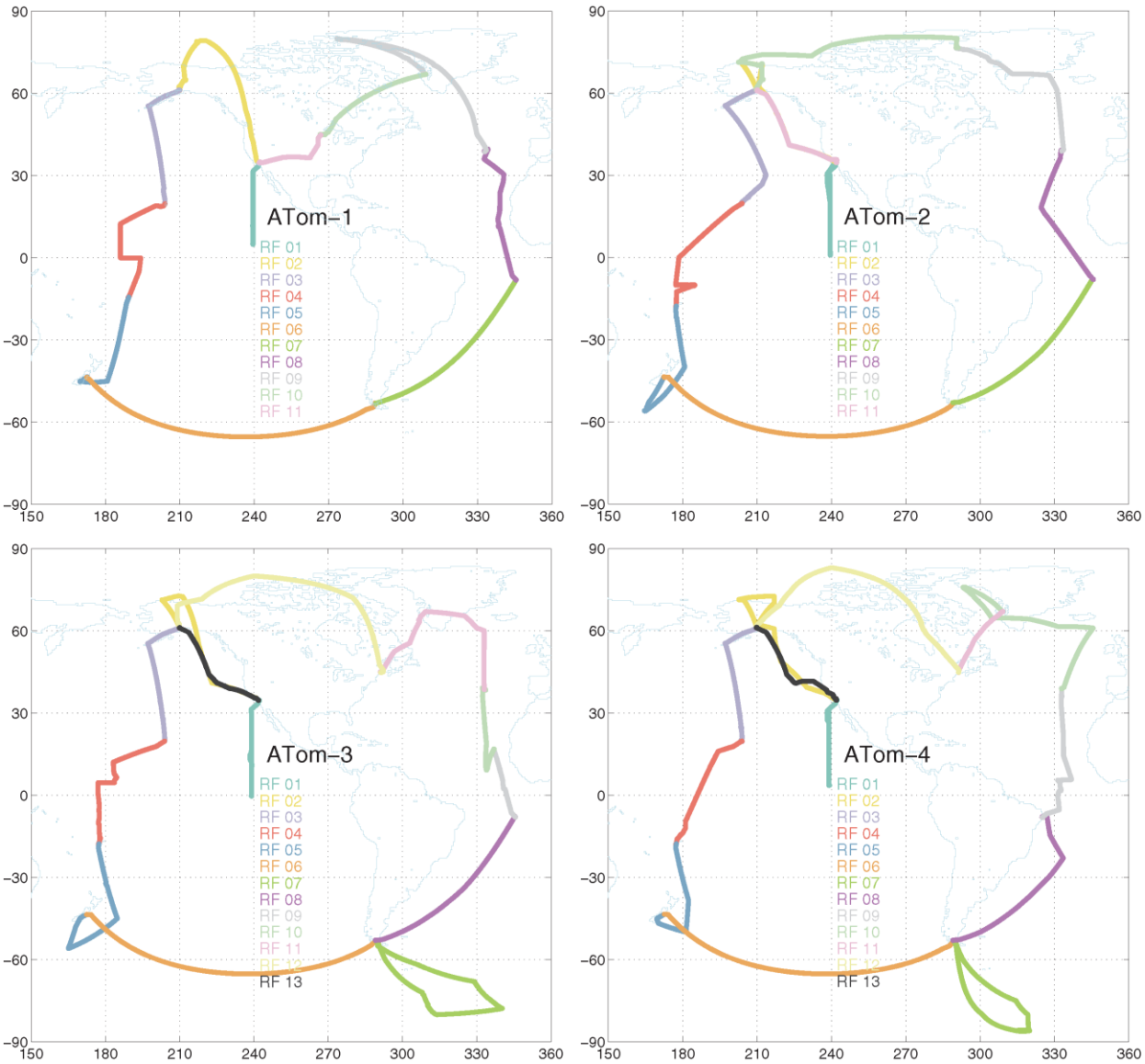

Figure S1. Map of ATom1234 flights, noting Research Flight number .

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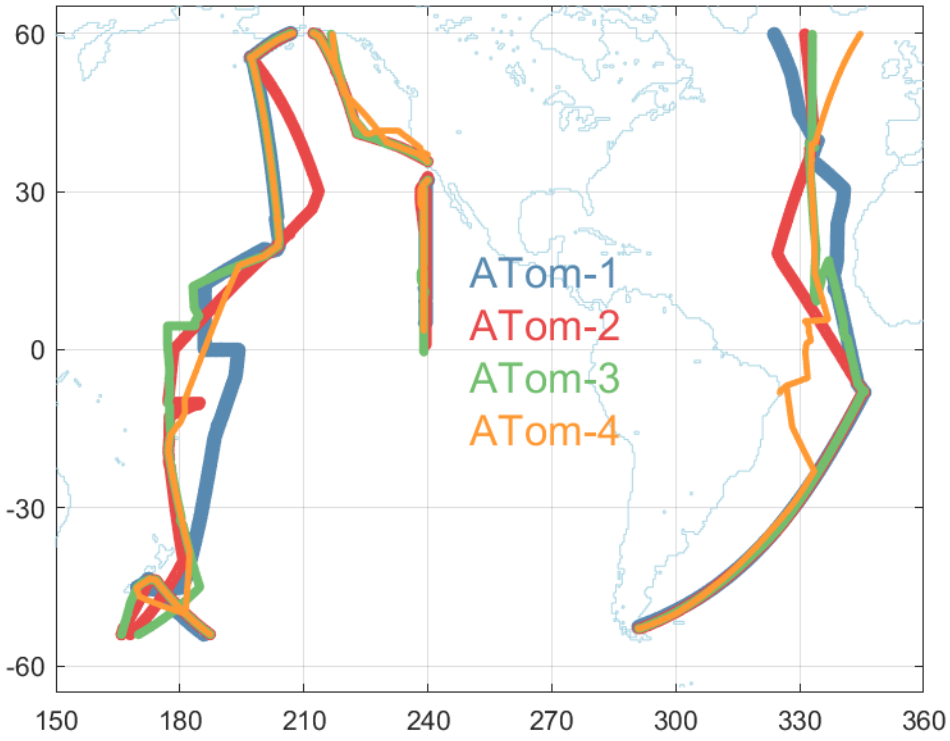

Figure S2. Map of the portion of ATom1234 flights included in the Pacific and Atlantic basin analysis. Flight tracks are plotted in successively thinner lines to see the overlap.

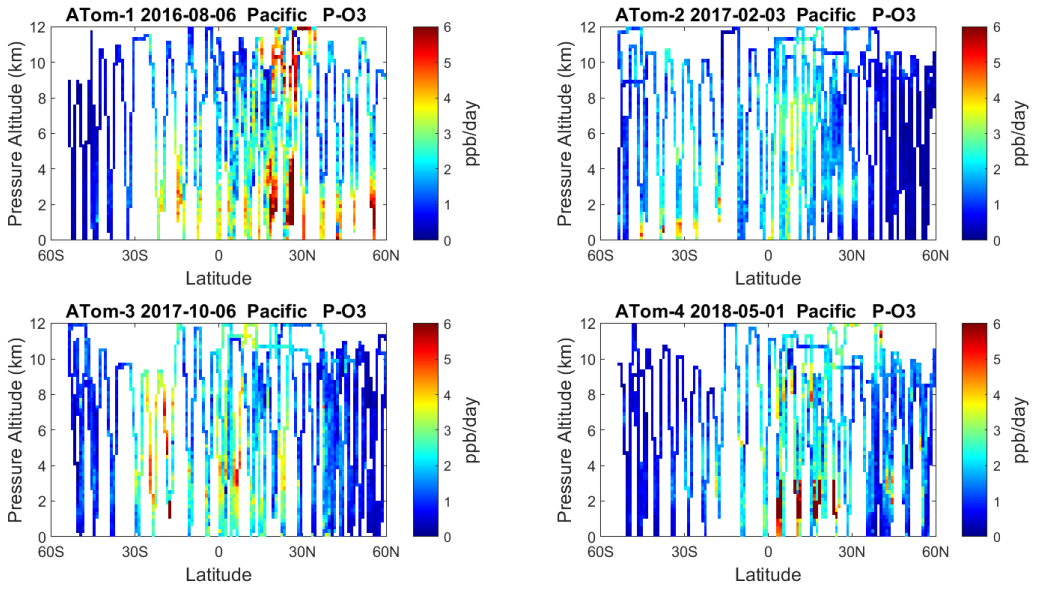

Figure S3. Altitude-Latitude profiling of P-O3 (ppb/day) in Pacific basin (54S-60N) for Atom-1234. All ATom 10s parcels are weighted by cosine (latitude).

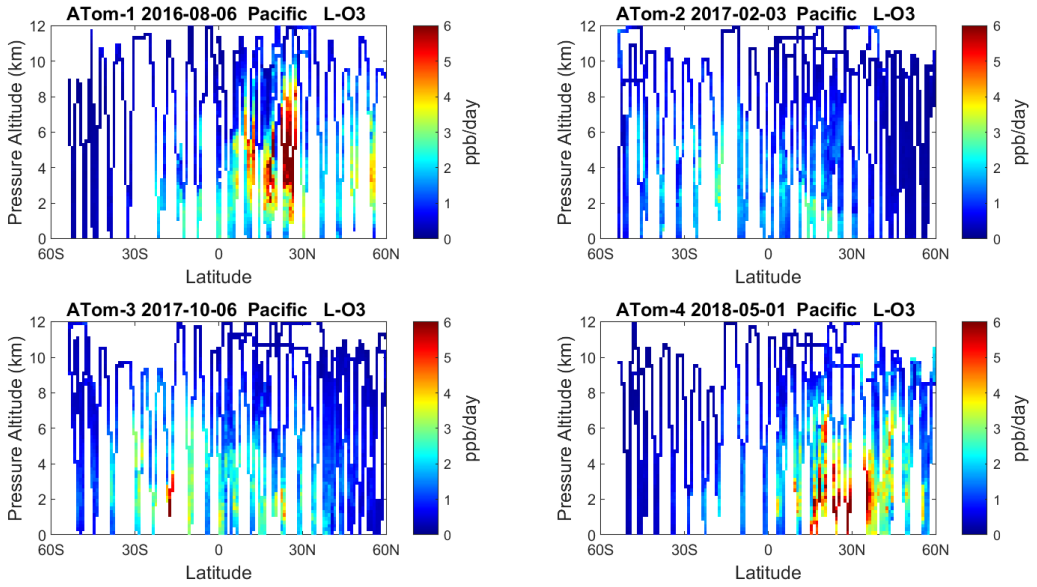

Figure S4. Altitude-Latitude profiling of L-O3 (ppb/day) in Pacific basin (54S-60N) for Atom-1234. All ATom 10s parcels are weighted by cosine (latitude).

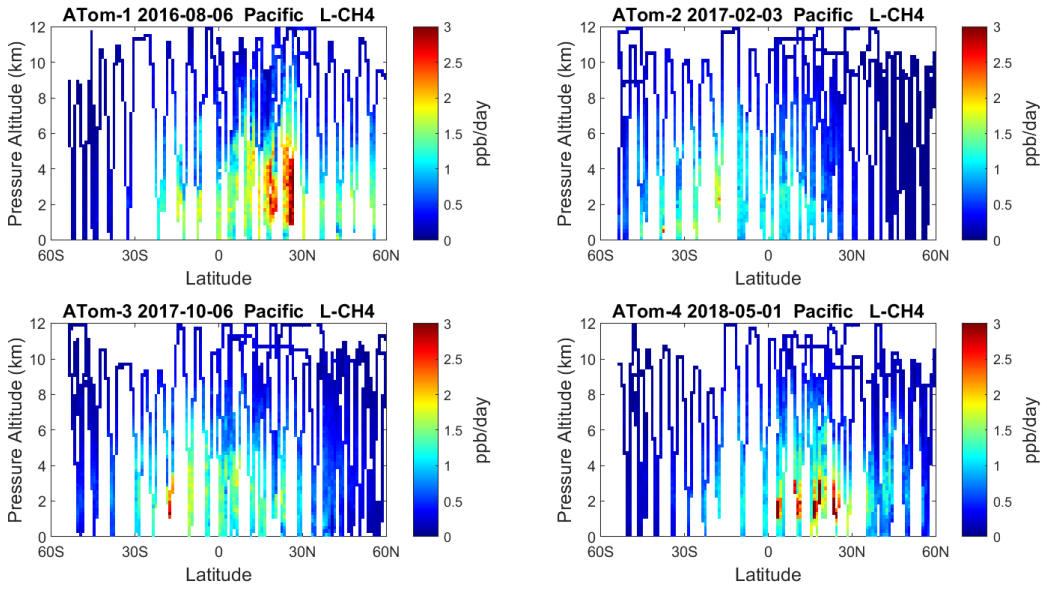

Figure S5. Altitude-Latitude profiling of L-CH4 (ppb/day) in Pacific basin (54S-60N) for Atom-1234. All ATom 10s parcels are weighted by cosine (latitude).

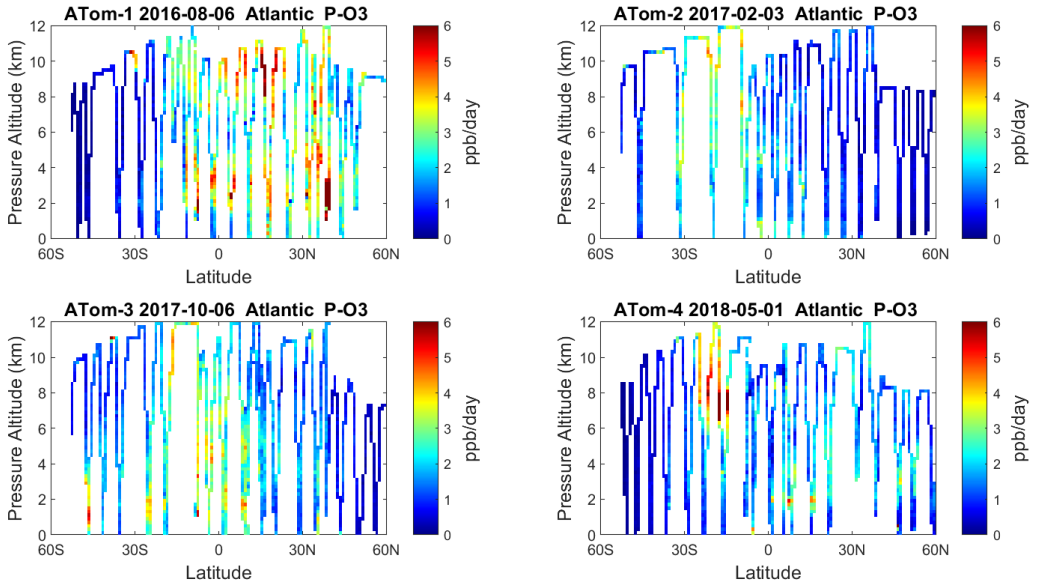

Figure S6. Altitude-Latitude profiling of P-O3 (ppb/day) in Atlantic basin (54S-60N) for Atom-1234. All ATom 10s parcels are weighted by cosine (latitude).

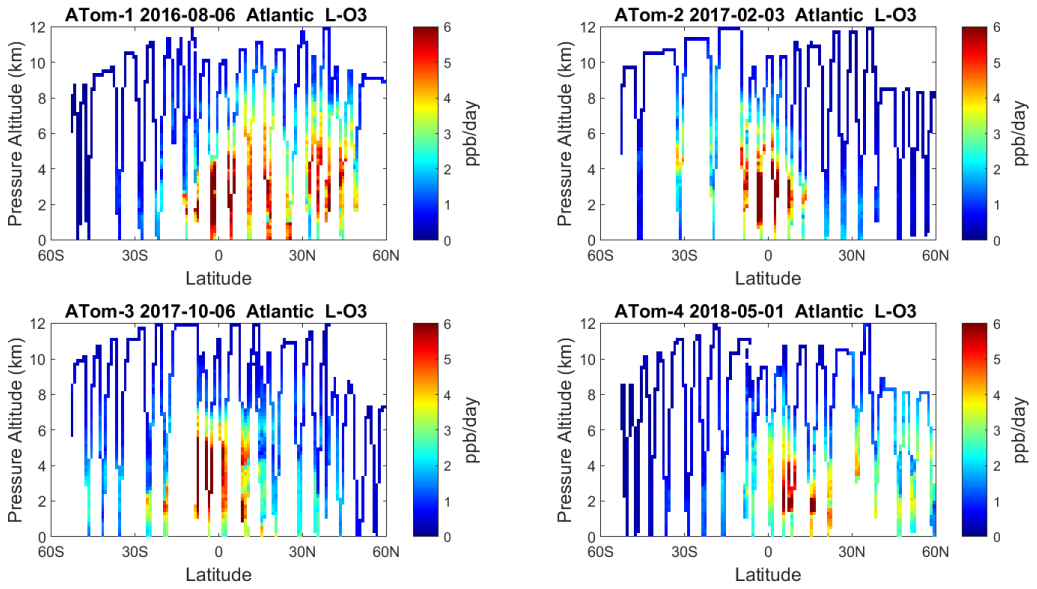

Figure S7. Altitude-Latitude profiling of L-O3 (ppb/day) in Atlantic basin (54S-60N) for Atom-1234. All ATom 10s parcels are weighted by cosine (latitude).

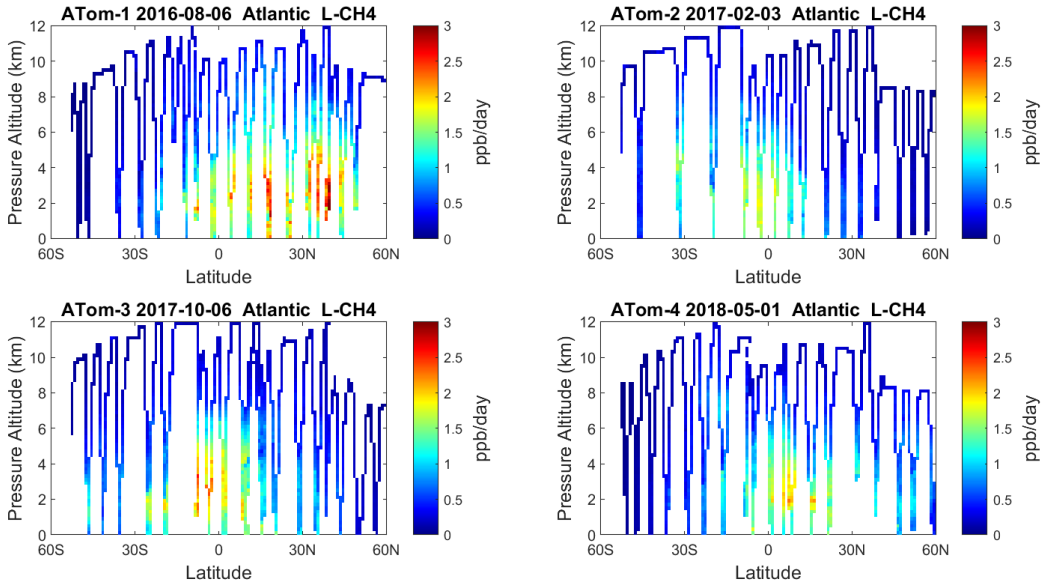

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Figure S8. Altitude-Latitude profiling of L-CH4 (ppb/day) in Atlantic basin (54S-60N) for Atom-1234. All ATom 10s parcels are weighted by cosine (latitude).

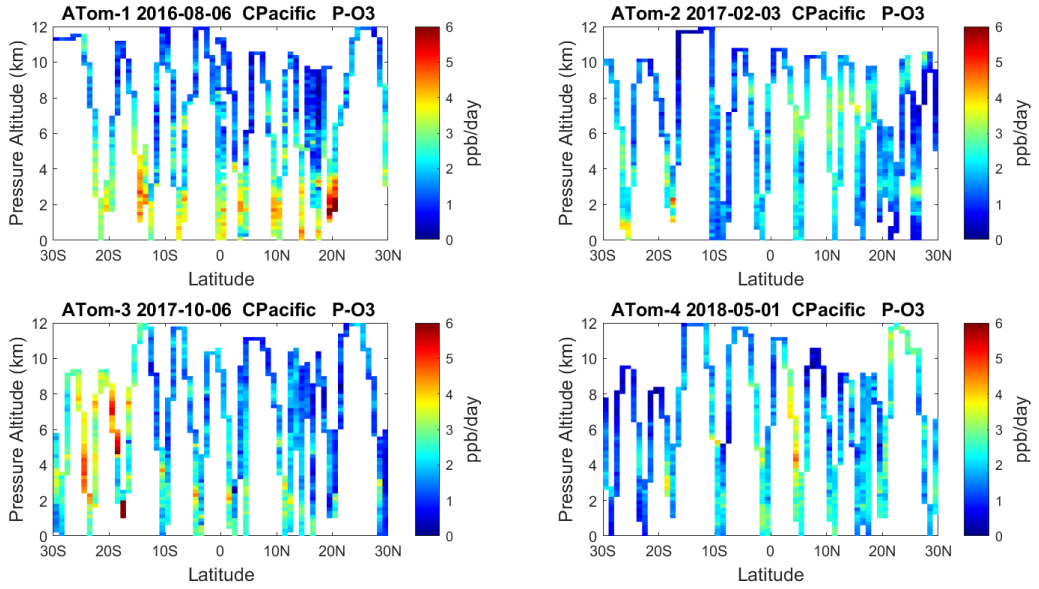

Figure S9. Altitude-Latitude profiling of P-O3 (ppb/day) in the tropical Central Pacific (30S-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

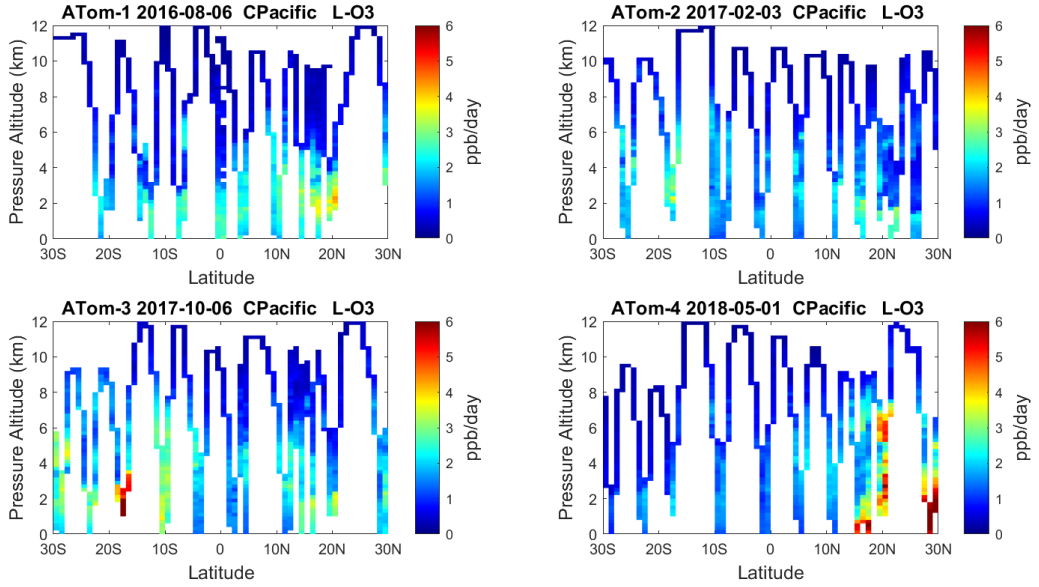

Figure S10. Altitude-Latitude profiling of L-O3 (ppb/day) in the tropical Central Pacific (30S-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

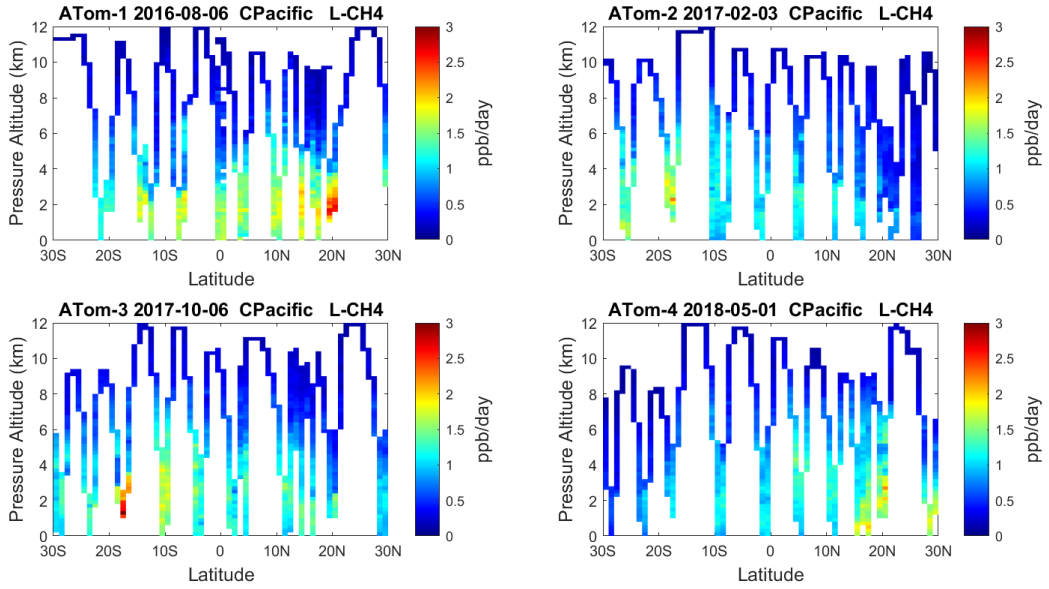

Figure S11. Altitude-Latitude profiling of L-CH4 (ppb/day) in the tropical Central Pacific (30S-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

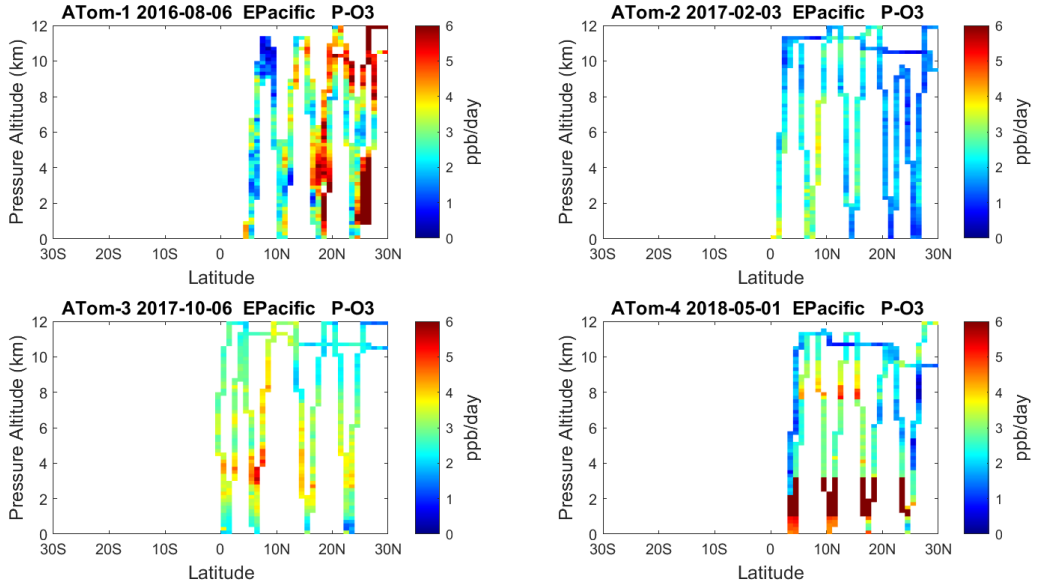

Figure S12. Altitude-Latitude profiling of P-O3 (ppb/day) in the tropical Eastern Pacific (0-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

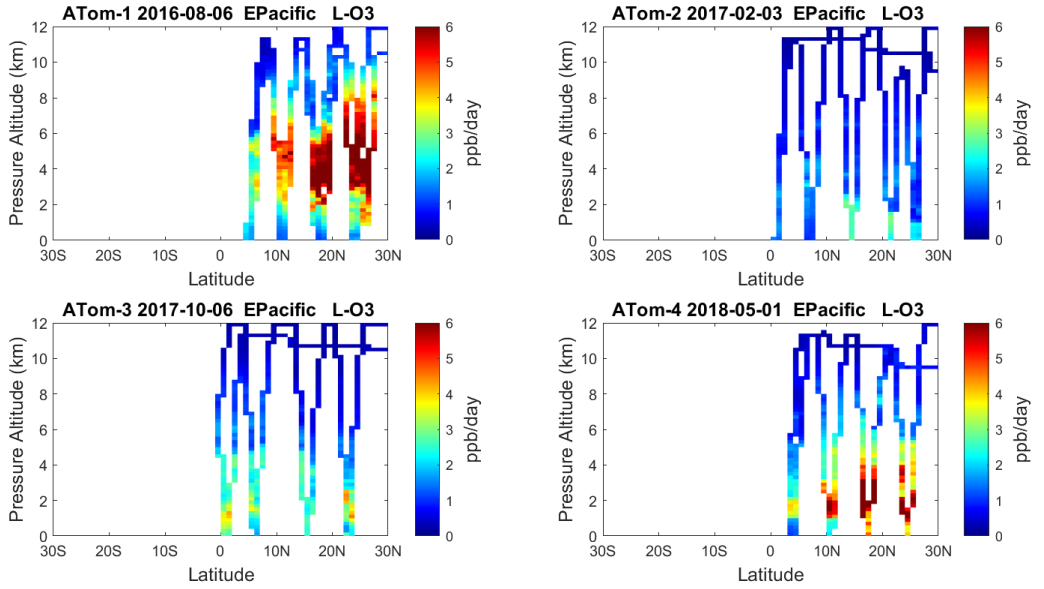

Figure S13. Altitude-Latitude profiling of L-O3 (ppb/day) in the tropical Eastern Pacific (0-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

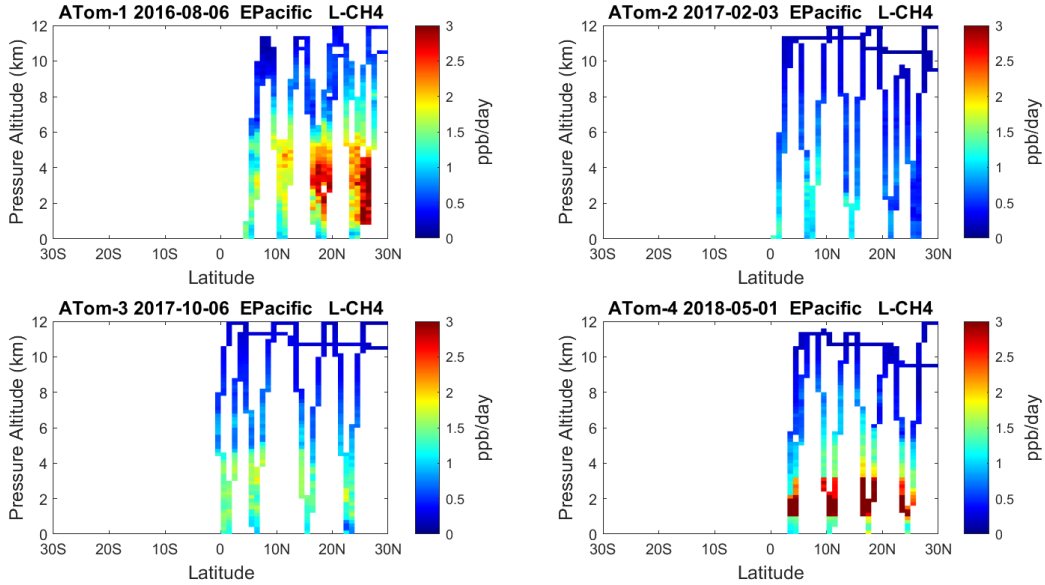

Figure S14. Altitude-Latitude profiling of L-CH4 (ppb/day) in the tropical Eastern Pacific (0-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

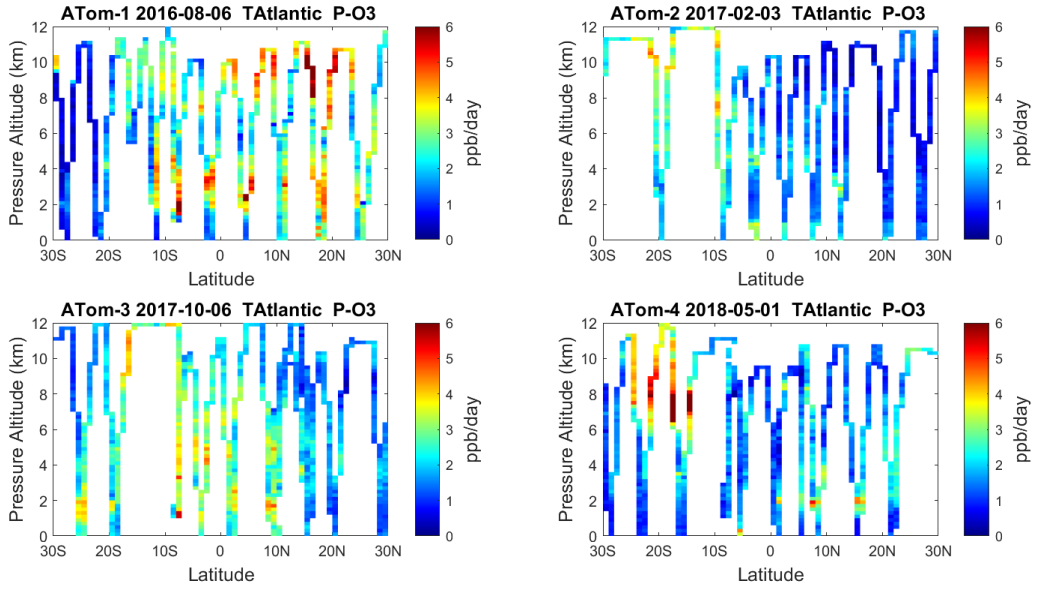

Figure S15. Altitude-Latitude profiling of P-O3 (ppb/day) in the tropical Atlantic (30S-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

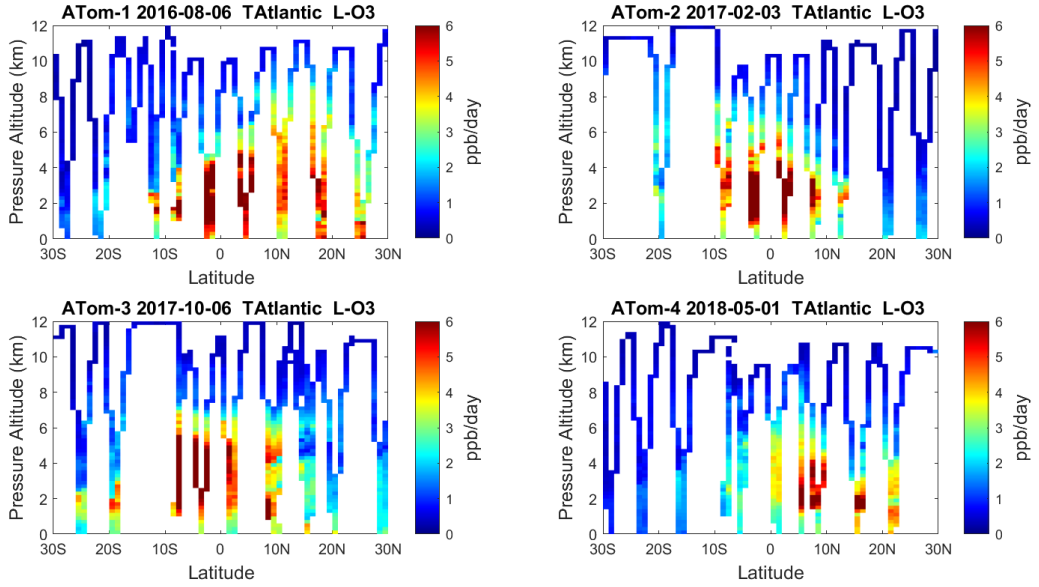

Figure S16. Altitude-Latitude profiling of L-O3 (ppb/day) in the tropical Atlantic (30S-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

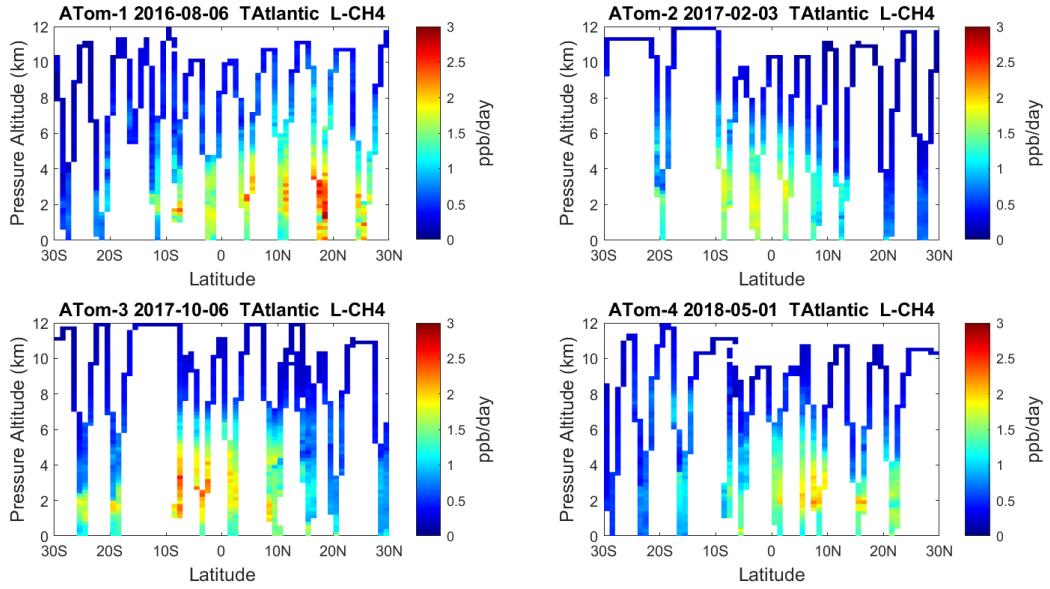

Figure S17. Altitude-Latitude profiling of L-CH4 (ppb/day) in the tropical Atlantic (30S-30N) sampled by ATom. All ATom 10s parcels are weighted by cosine (latitude).

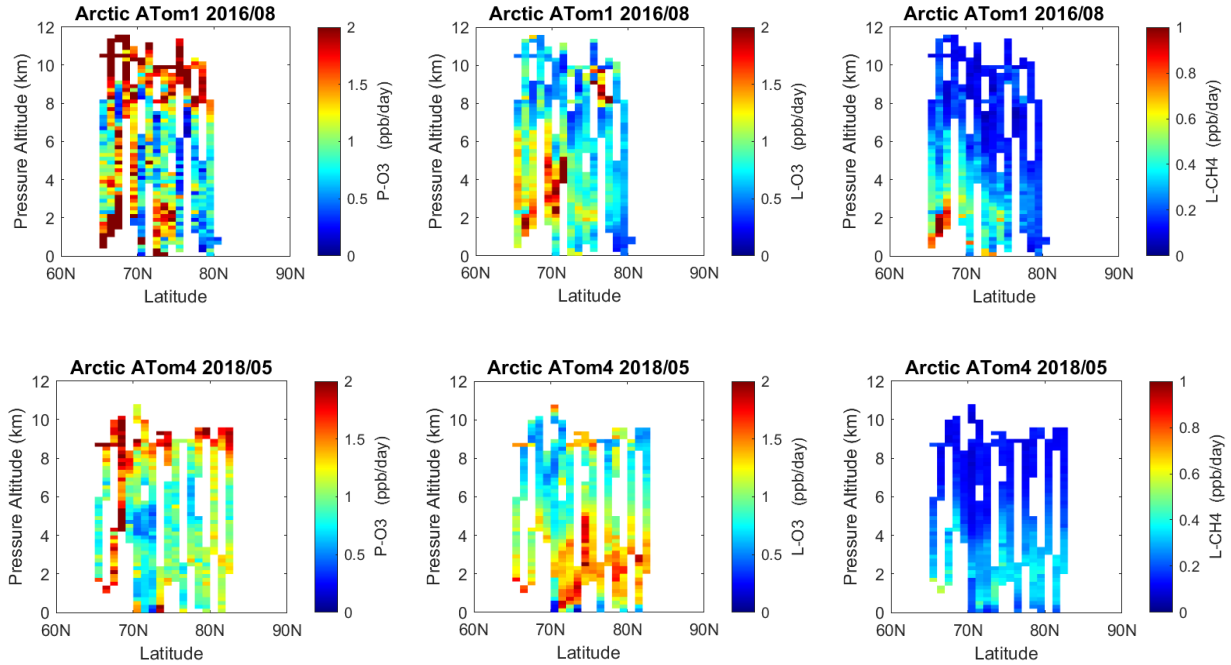

Figure S18. Altitude-Latitude profiling of the 3 reactivities (P-O3, L-O3, L-CH4, ppb/day) over the Arctic on ATom1 (top) and ATom4 (bottom). The noontime solar zenith angles on ATom2 and ATom4 were so large that reactivities are small and not shown. Note the color bars have a much range than in the similar Pacific and Atlantic basin plots. Troposphere only parcels, with stratosphere defined as (H2O < 30 ppm) and (O3 > 80 ppb) and (CO < 120 ppb).

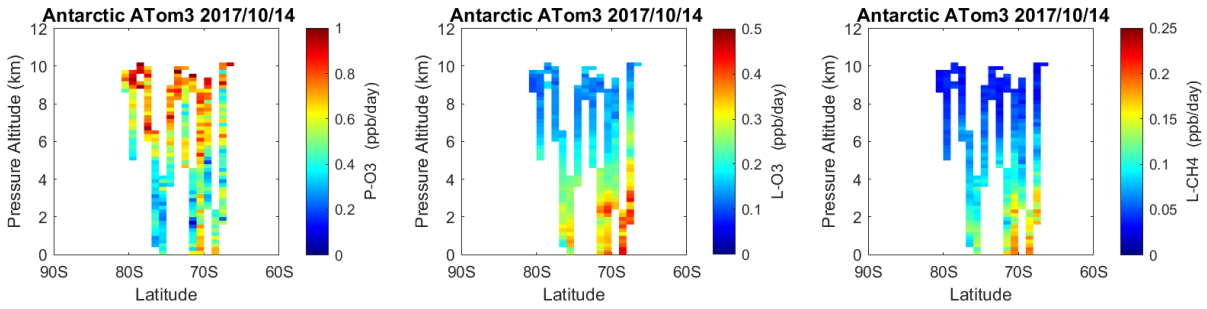

Figure S19. Altitude-Latitude profiling of the 3 reactivities (P-O3, L-O3, L-CH4, ppb/day) over Antarctica on ATom3. For ATom4 (2018/05/09) noontime solar zenith angles were large, and the very small reactivities are not shown. Note the color bars have a smaller range than in the similar Pacific and Atlantic basin plots. Troposphere only parcels, with stratosphere defined as (H2O < 30 ppm) and (O3 > 80 ppb) and (CO < 120 ppb).

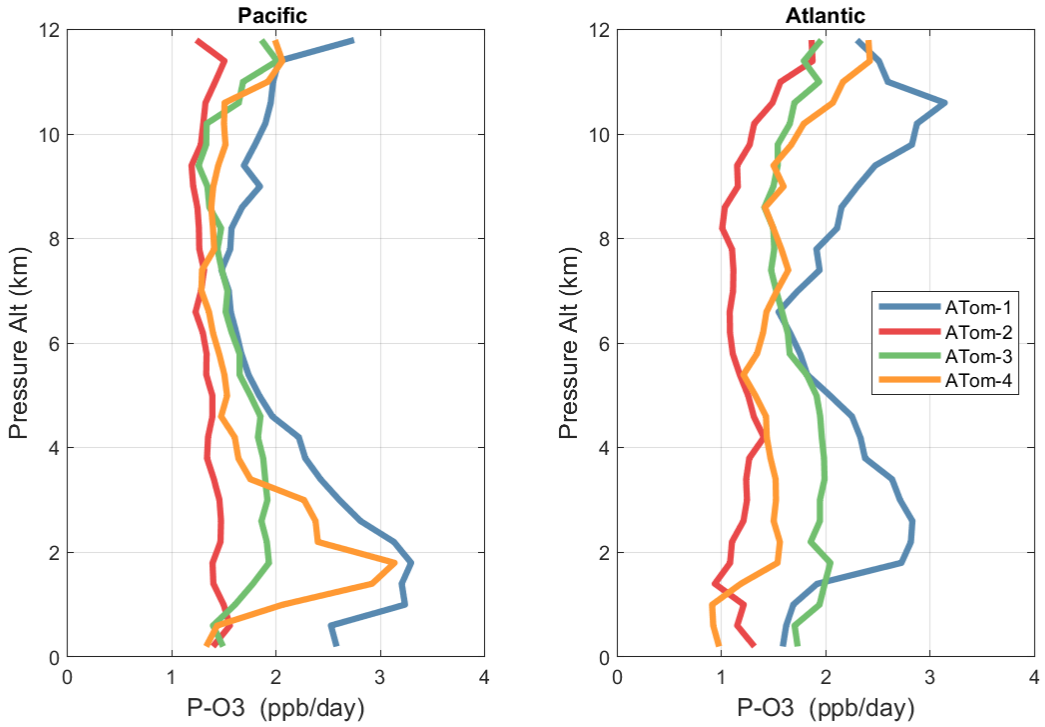

Figure S20. Mean altitude profile of P-O3 (ppb/day) over the Pacific and Atlantic basins (54S-60N) for ATom1234.

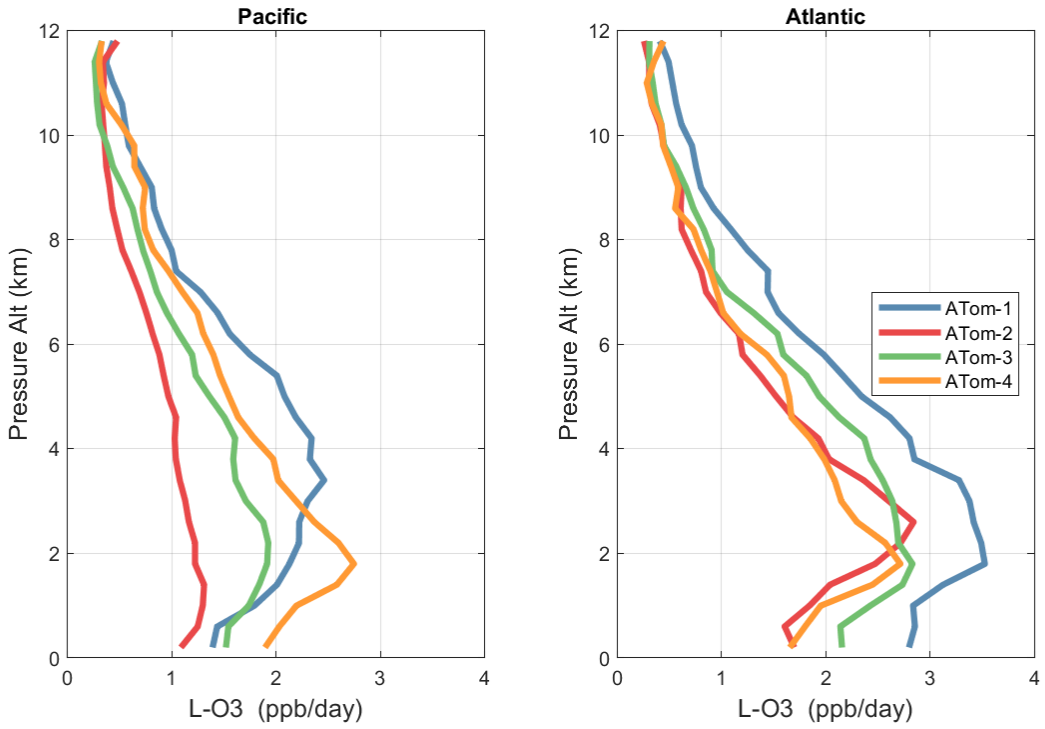

Figure S21. Mean altitude profile of L-O3 (ppb/day) over the Pacific and Atlantic basins (54S-60N) for ATom1234.

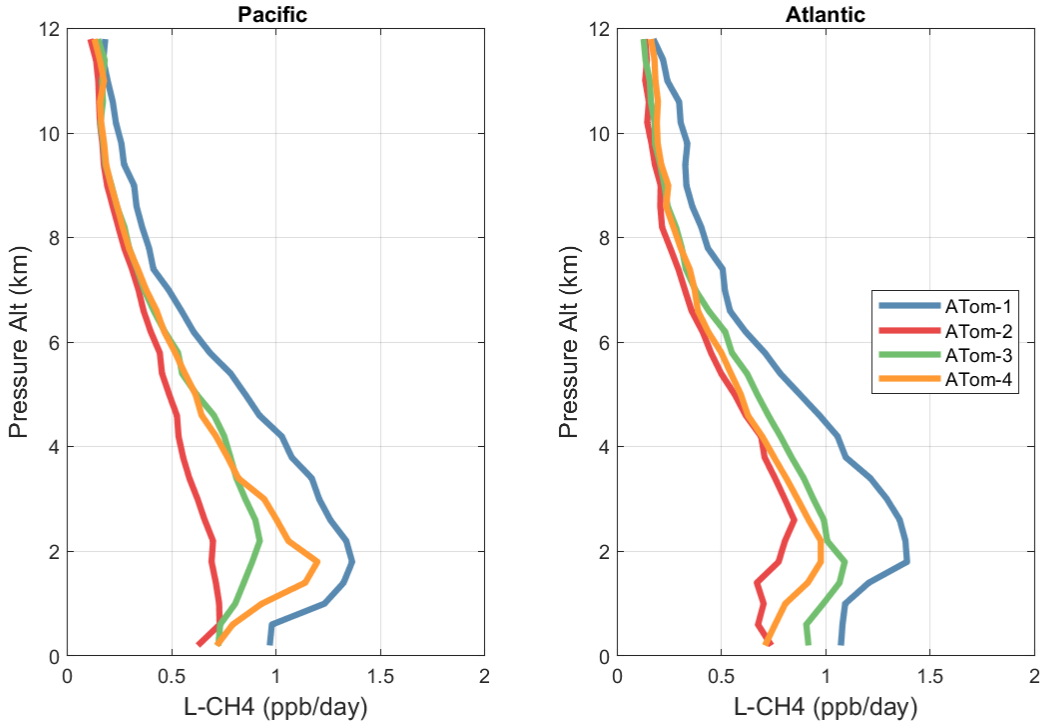

Figure S22. Mean altitude profile of L-CH4 (ppb/day) over the Pacific and Atlantic basins (54S-60N) for ATom1234.

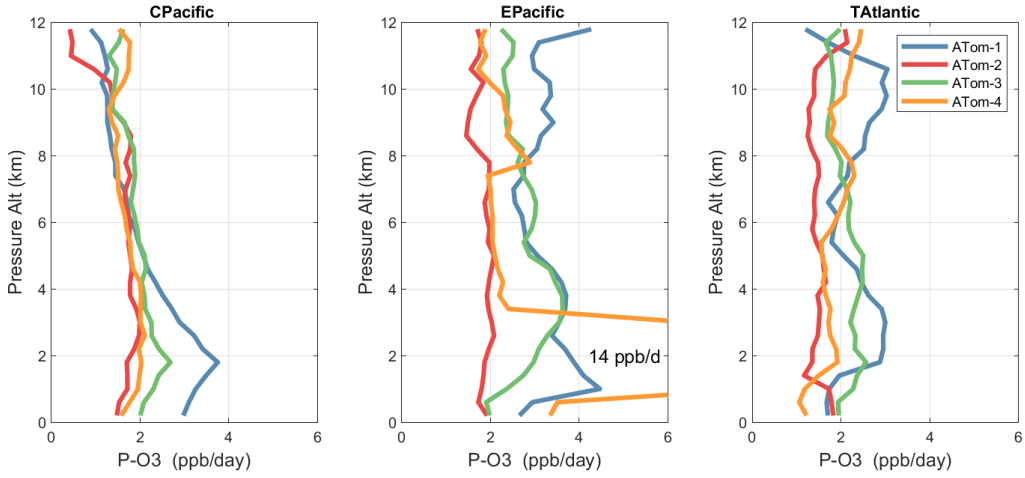

Figure S23. Mean altitude profile of P-O3 (ppb/day) over the 3 tropical basins for ATom1234.

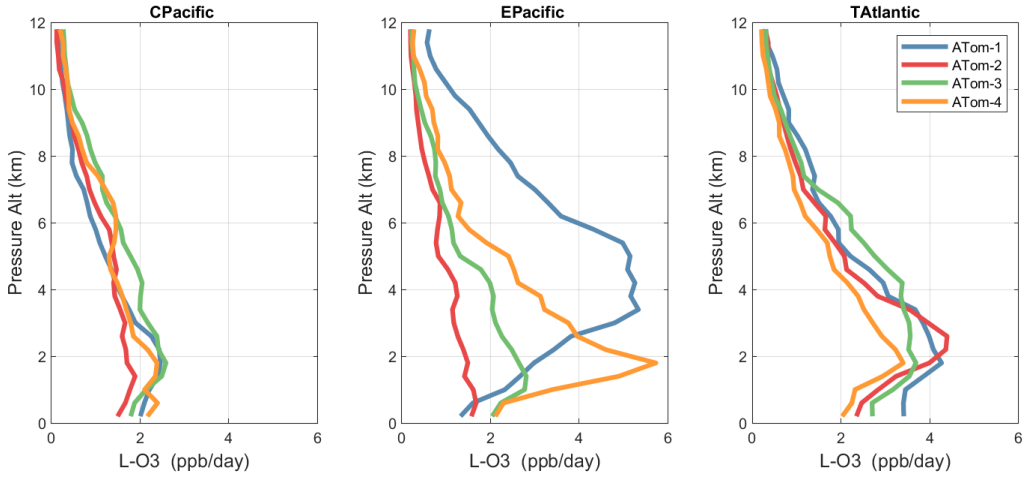

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Figure S24. Mean altitude profile of L-O3 (ppb/day) over the 3 tropical basins for ATom1234.

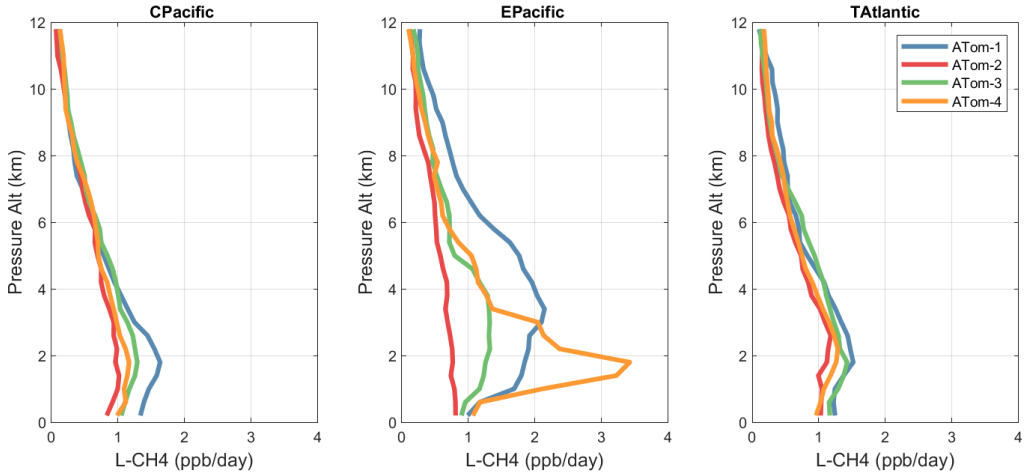

Figure S25. Mean altitude profile of L-CH4 (ppb/day) over the 3 tropical basins for ATom1234.

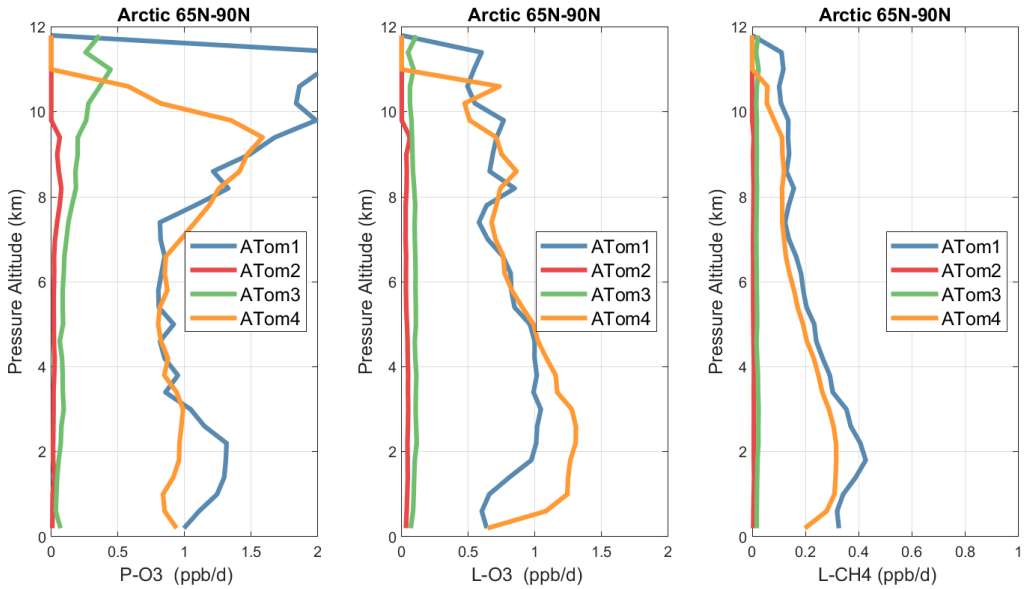

Figure S26. Mean altitude profile of the 3 reactivities (P-O3, L-O3, L-CH4, ppb/day) over the Arctic (65N-90N) for ATom1234. All ATom 10s parcels are weighted equally. Troposphere only parcels, with stratosphere defined as (H2O < 30 ppm) and (O3 > 80 ppb) and (CO < 120 ppb).

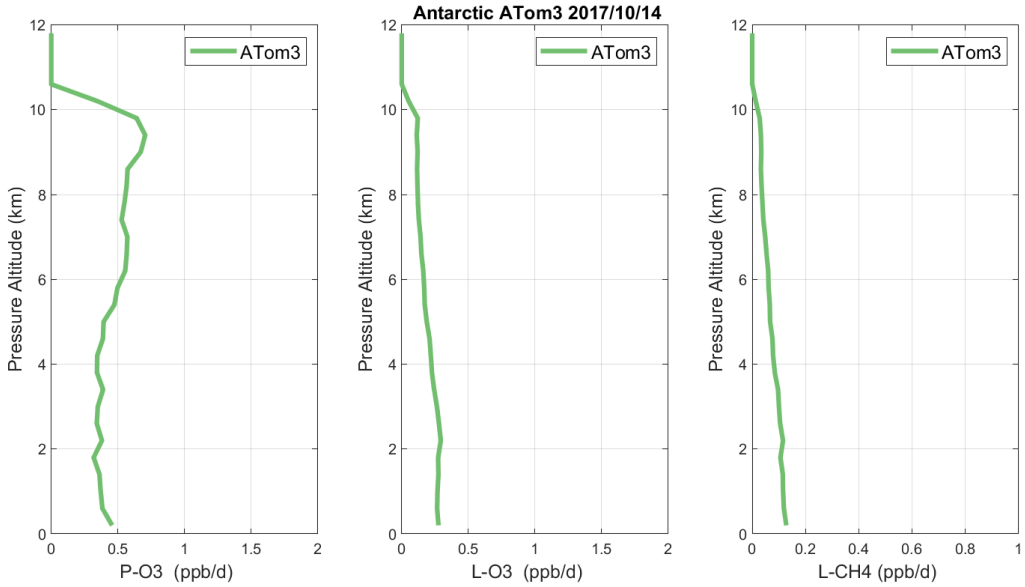

Figure S27. Mean altitude profile of the 3 reactivities (P-O3, L-O3, L-CH4, ppb/day) over Antarctica on ATom3. For ATom4 (2018/05/09) noontime solar zenith angles were large, and the very small reactivities are not shown. All ATom 10s parcels are weighted equally. Troposphere only parcels, with stratosphere defined as (H2O < 30 ppm) and (O3 > 80 ppb) and (CO < 120 ppb).

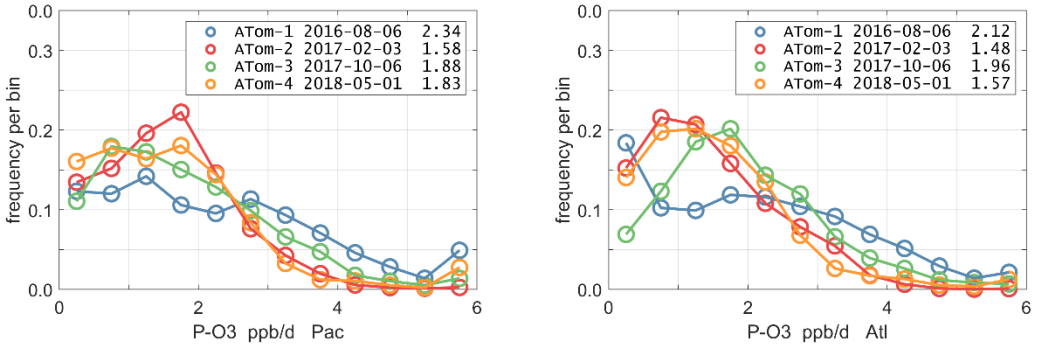

Figure S28. Probability Density of P-O3 (ppb/day) in Pacific and Atlantic basins (54S-60N) for Atom-1234.

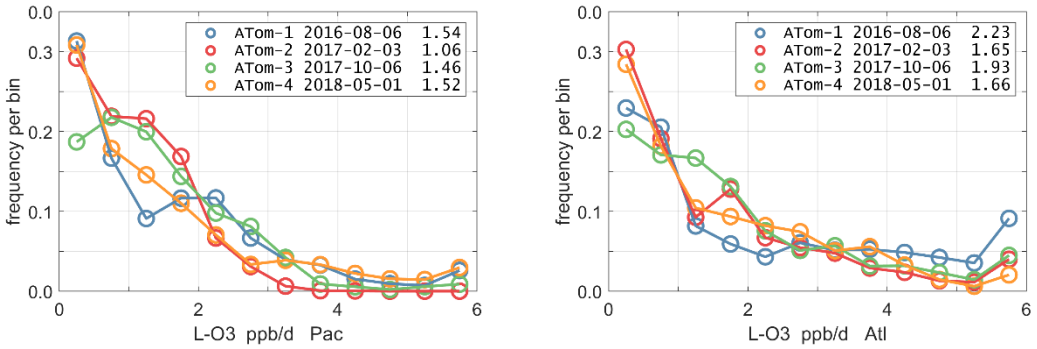

Figure S29. Probability Density of L-O3 (ppb/day) in Pacific and Atlantic basins (54S-60N) for Atom-1234.

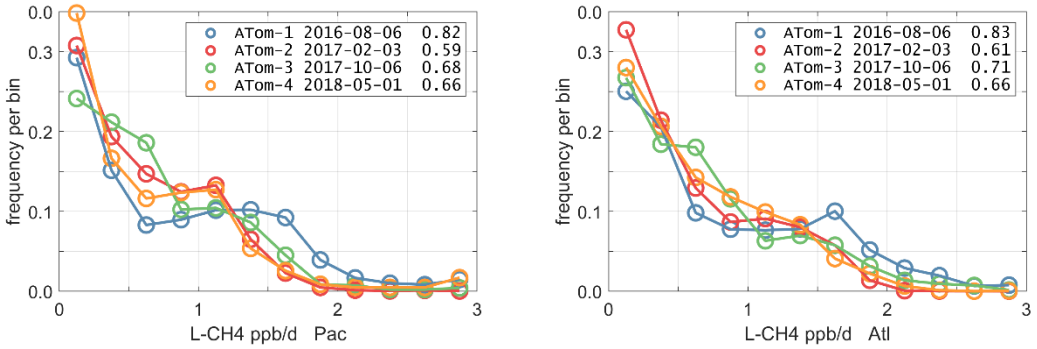

Figure S30. Probability Density of L-CH4 (ppb/day) in Pacific and Atlantic basins (54S-60N) for Atom-1234.

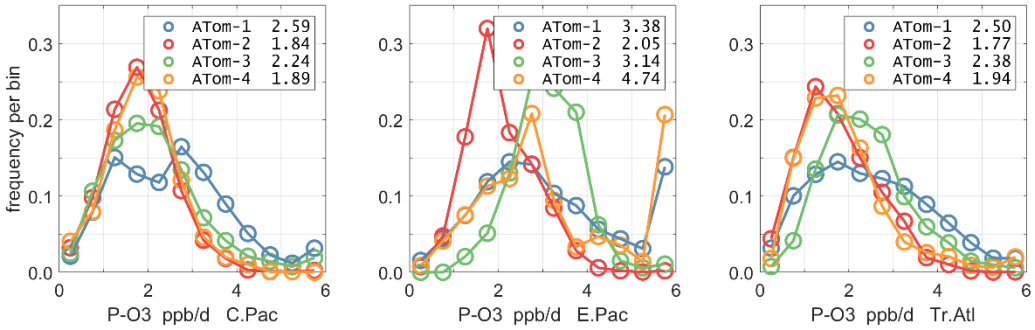

Figure S31. Probability Density of P-O3 (ppb/day) in the 3 tropical basins (30S-30N) for Atom-1234.

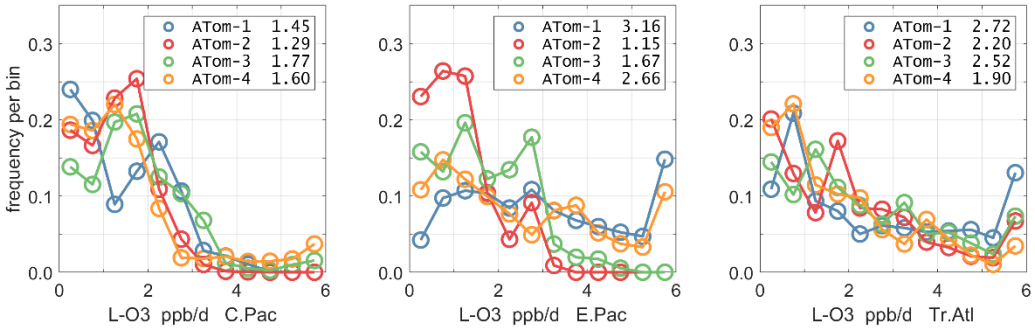

Figure S32. Probability Density of L-O3 (ppb/day) in the 3 tropical basins (30S-30N) for Atom-1234.

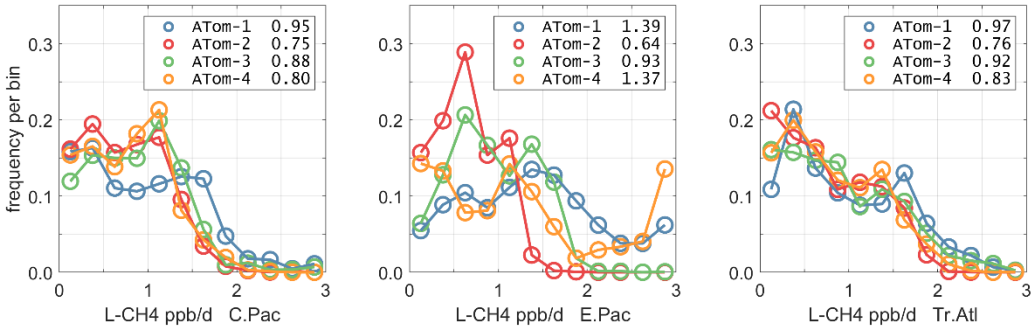

Figure S33. Probability Density of L-CH4 (ppb/day) in the 3 tropical basins (30S-30N) for Atom-1234.

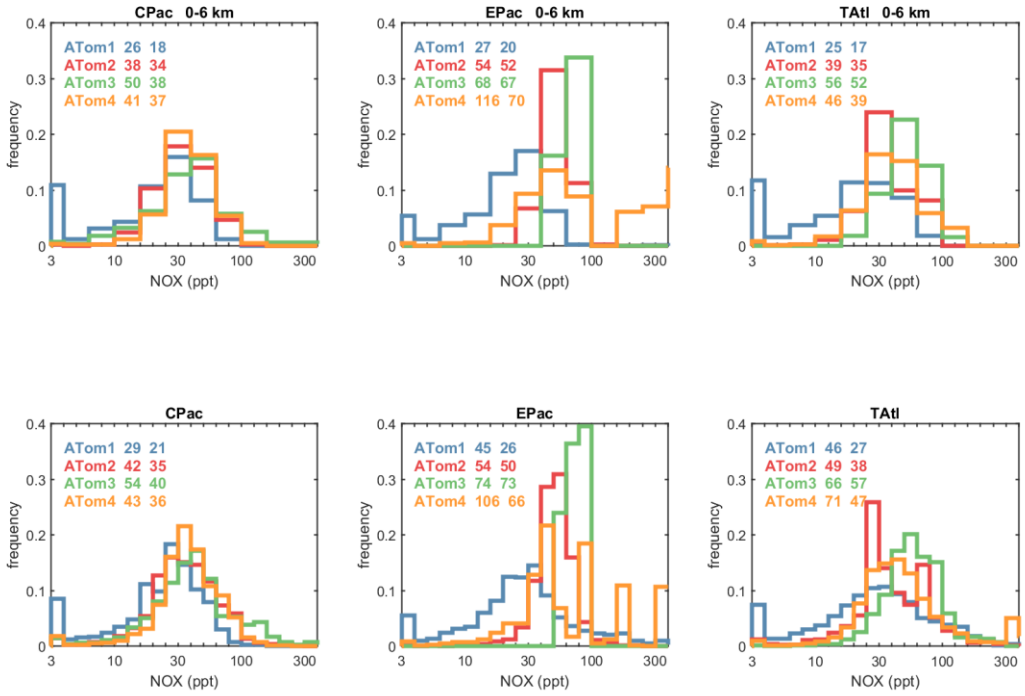

Figure S34. Probability Density of log10(NOx, ppt) in the 3 tropical basins (30S-30N) for Atom-1234. The standard weighting of ATom 10s air parcels is used. The upper panel shows 0-6 km pressure altitude where most of the chemical reactivity is located; while the lower panel shows the full troposphere, approximately 0-12 km. The color coding in the legend identifies the 4 ATom deployments. The numbers in the legend are, successively, the mean value of NOx and the mean value of the log10(NOx), both in ppt.

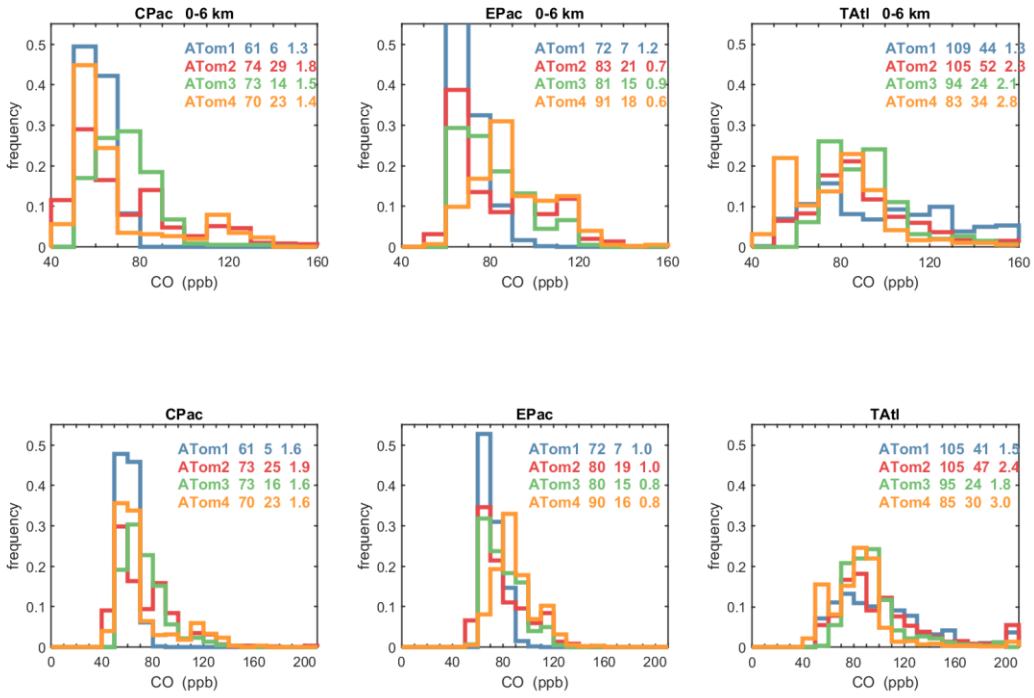

Figure S35. Probability Density of CO (ppb) in the 3 tropical basins (30S-30N) for Atom-1234. The standard weighting of ATom 10s air parcels is used. The upper panel shows 0-6 km pressure altitude where most of the chemical reactivity is located; while the lower panel shows the full troposphere, approximately 0-12 km. The color coding in the legend identifies the 4 ATom deployments. The numbers in the legend are, successively, the mean value, standard deviation, and skewness.

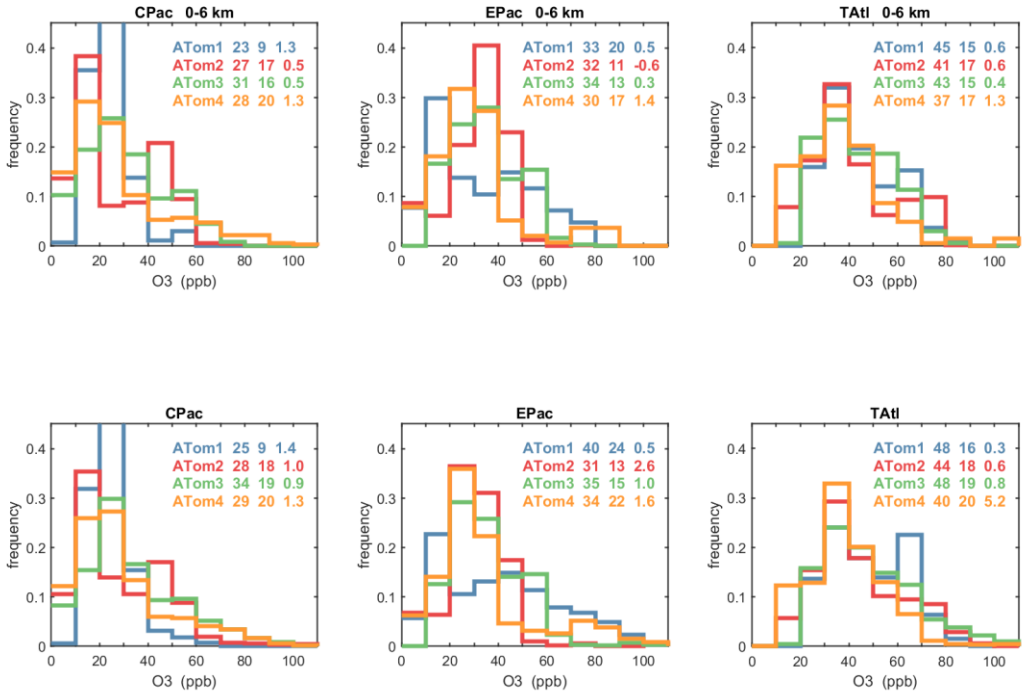

Figure S36. Probability Density of O3 (ppb) in the 3 tropical basins (30S-30N) for Atom-1234. The standard weighting of ATom 10s air parcels is used. The upper panel shows 0-6 km pressure altitude where most of the chemical reactivity is located; while the lower panel shows the full troposphere, approximately 0-12 km. The color coding in the legend identifies the 4 ATom deployments. The numbers in the legend are, successively, the mean value, standard deviation, and skewness.

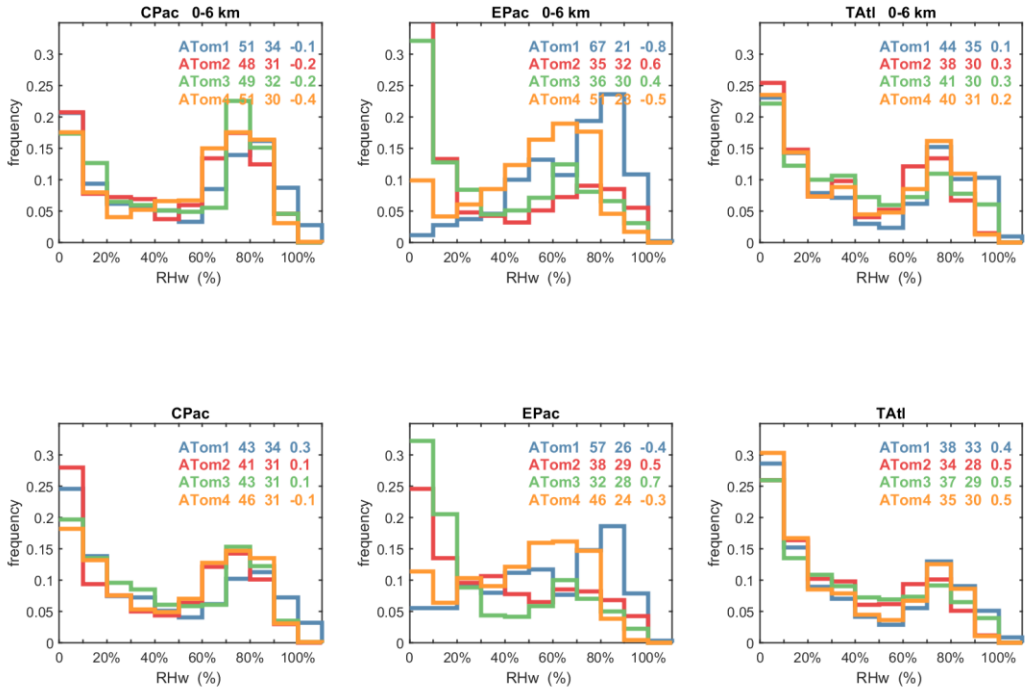

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Figure S37. Probability Density of the relative humidity over liquid water (RHw, %) in the 3 tropical basins (30S-30N) for Atom-1234. The standard weighting of ATom 10s air parcels is used. The upper panel shows 0-6 km pressure altitude where most of the chemical reactivity is located; while the lower panel shows the full troposphere, approximately 0-12 km. The color coding in the legend identifies the 4
150 ATom deployments. The numbers in the legend are, successively, the mean value, standard deviation, and skewness.

Table S1. First-order sensitivities of the reactivities (R) with respect to the initial value of key species (X), $S \equiv \partial[\ln(R)]/\partial[\ln(X)]$ (%/%) are calculated with a perturbation of 10%. Results are shown separately for the 4 deployments (ATom-1234) and the Pacific and Atlantic basins (54°S to 60°N). The average and standard deviation are shown on the right. For H_2O and T, results include only ATom-1.

P-O3		**ATom-1**			**ATom-2**			**ATom-3**			**ATom-4**			**avg**	*std*
		Pacific	*Atlantic*		*Pacific*	*Atlantic*		*Pacific*	*Atlantic*		*Pacific*	*Atlantic*			
NOx	%/%	**0.19**	**0.22**		**0.18**	**0.22**		**0.19**	**0.23**		**0.20**	**0.22**		**0.21**	*0.02*
O3	%/%	**-0.53**	**-0.60**		**-0.53**	**-0.61**		**-0.47**	**-0.53**		**-0.52**	**-0.56**		**-0.54**	*0.04*
CH4	%/%	**0.16**	**0.18**		**0.14**	**0.16**		**0.14**	**0.15**		**0.14**	**0.16**		**0.15**	*0.01*
CO	%/%	0.06	0.09		0.05	0.07		0.07	0.06		0.07	0.08		0.07	*0.01*
H2O	%/%	**0.14**	**0.15**											**0.14**	*0.00*
HCHO	%/%	0.02	0.02		0.04	0.03		0.03	0.05		0.02	0.02		0.03	*0.01*
H2O2	%/%	0.03	0.04		0.03	0.03		0.03	0.02		0.02	0.04		0.03	*0.01*
PAN	%/%	0.06	0.05		0.03	0.05		0.07	0.06		0.06	0.04		0.05	*0.01*
HNO3	%/%	0.05	0.07		0.07	0.08		0.06	0.07		0.04	0.06		0.06	*0.01*
HNO4	%/%	0.05	0.07		0.07	0.08		0.06	0.06		0.04	0.06		0.06	*0.01*
MeOOH	%/%	**0.09**	0.06		**0.10**	0.07		**0.12**	0.06		**0.12**	0.05		0.08	*0.03*
C2H6	%/%	0.00	0.00		0.00	0.00		0.00	0.00		0.00	0.00		0.00	*0.00*
Alkane	%/%	0.00	0.00		0.00	0.00		0.00	0.00		0.00	0.00		0.00	*0.00*

L-O3		**ATom-1**			**ATom-2**			**ATom-3**			**ATom-4**			**avg**	*std*
		Pacific	*Atlantic*		*Pacific*	*Atlantic*		*Pacific*	*Atlantic*		*Pacific*	*Atlantic*			
NOx	%/%	0.03	0.02		0.05	0.03		0.05	0.04		0.04	0.03		0.04	*0.01*
O3	%/%	**0.23**	**0.28**		**0.25**	**0.29**		**0.22**	**0.26**		**0.21**	**0.23**		**0.25**	*0.03*
CH4	%/%	-0.03	-0.03		-0.01	-0.03		-0.02	-0.02		-0.02	-0.02		-0.02	*0.01*
CO	%/%	0.00	0.00		0.00	0.01		0.00	0.00		0.01	0.00		0.00	*0.00*
H2O	%/%	**0.49**	**0.47**											**0.48**	*0.01*
HCHO	%/%	0.01	0.02		0.03	0.03		0.03	0.03		0.02	0.03		0.02	*0.01*
H2O2	%/%	0.04	0.04		0.03	0.03		0.03	0.02		0.04	0.03		0.03	*0.01*
PAN	%/%	0.01	0.01		0.01	0.01		0.01	0.01		0.01	0.01		0.01	*0.00*
HNO3	%/%	0.01	0.01		0.01	0.01		0.00	0.00		0.00	0.01		0.01	*0.00*
HNO4	%/%	0.01	0.01		0.01	0.01		0.00	0.00		0.00	0.01		0.01	*0.00*
MeOOH	%/%	0.02	0.01		0.02	0.01		0.03	0.03		0.02	0.01		0.02	*0.01*
C2H6	%/%	0.00	0.00		0.00	0.00		0.00	0.00		0.00	0.00		0.00	*0.00*
Alkane	%/%	0.00	0.00		0.00	0.00		0.00	0.00		0.00	0.00		0.00	*0.00*

L-CH4		**ATom-1**			**ATom-2**			**ATom-3**			**ATom-4**			**avg**	*std*
		Pacific	*Atlantic*		*Pacific*	*Atlantic*		*Pacific*	*Atlantic*		*Pacific*	*Atlantic*			
NOx	%/%	**0.09**	0.08		0.08	0.08		**0.10**	0.09		**0.10**	**0.10**		**0.09**	*0.01*
O3	%/%	**0.39**	**0.31**		**0.40**	**0.34**		**0.39**	**0.34**		**0.38**	**0.35**		**0.36**	*0.03*
CH4	%/%	**0.69**	**0.73**		**0.68**	**0.73**		**0.70**	**0.73**		**0.67**	**0.72**		**0.71**	*0.02*
CO	%/%	**-0.34**	**-0.40**		**-0.36**	**-0.40**		**-0.37**	**-0.38**		**-0.37**	**-0.39**		**-0.38**	*0.02*
H2O	%/%	**0.39**	**0.36**											**0.38**	*0.01*

HCHO	%/%	0.00	0.01		0.00	0.01		0.01	0.01		0.01	0.01		0.01	*0.00*
H2O2	%/%	0.06	0.06		0.05	0.06		0.05	0.04		0.04	0.06		0.05	*0.01*
PAN	%/%	0.03	0.02		0.02	0.02		0.01	0.03		0.03	0.02		0.02	*0.01*
HNO3	%/%	0.02	0.02		0.02	0.02		0.01	0.03		0.01	0.01		0.02	*0.01*
HNO4	%/%	0.02	0.02		0.02	0.02		0.00	0.02		0.02	0.01		0.02	*0.01*
MeOOH	%/%	-0.04	-0.02		-0.04	-0.03		-0.02	-0.03		-0.04	-0.03		-0.03	*0.01*
C2H6	%/%	-0.01	0.00		0.00	0.00		0.00	0.00		0.00	0.00		0.00	*0.00*
Alkane	%/%	0.00	0.00		0.00	0.00		0.00	0.00		0.00	0.00		0.00	*0.00*

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Table S2. Second-order, quadratic sensitivities of the 3 reactivities relative to critical species (S > 0.1) calculated with 20% and 10% perturbations, shown as $S_{+20\%}$ / $S_{+10\%}$. Results are shown for ATom-1 and both Pacific and Atlantic basins (54°S-60°N). There is little evidence of quadratic curvature in the sensitivities for perturbations <20%.

	Pacific			Atlantic		
species	P-O3	L-O3	L-CH4	P-O3	L-O3	L-CH4
NOx	+.20 / +.19	+.03 / +.03	+.10 / +.09	+.23 / +.22	+.03 / +.02	+.09 / +.08
O3	-.54 / -.53	+.23 / +.23	+.39 / +.39	-.61 / -.60	+.28 / +.28	+.33 / +.31
CH4	+.16 / +.16	-.03 / -.03	+.68 / +.69	+.18 / +.18	-.03 / -.03	+.72 / +.73
CO	+.06 / +.06	.00 / .00	-.35 / -.34	+.09 / +.09	.00 / .00	-.41 / -.40

Table S3. Second-order, cross-term sensitivities of the 3 reactivities (P-O3, L-O3, L-CH4) relative to critical species (NOx, O_3, CH_4, CO) calculated in pair combinations. Results here are from ATom-1 and the Pacific and Atlantic basins (54°S-60°N). The single +10% sensitivities calculated for individual species, S(X+10%), are shown in italics along the diagonal. The doubly signed (i.e., ++ or --) off-diagonal values represent the additional change in sensitivity due to the coupling of the two species: S(X+10% & Y+10%) – S(X+10%) – S(Y+10%). When the 2nd-order is small, the coupled perturbation is simply the sum of the two individual ones; but when it is large, the coupling of perturbations becomes important.

	Pacific					**Atlantic**			
P-O3	NOx	O_3	CH_4	CO		NOx	O_3	CH_4	CO
NOx	*+0.19*	**++0.08**	**++0.05**	**++0.10**		*+0.22*	**++0.08**	**++0.03**	**++0.09**
O_3		*-0.53*	**++0.08**	**++0.04**			*-0.60*	**++0.07**	**++0.06**
CH_4			*+0.16*	**--0.07**				*+0.18*	**--0.06**
CO				*+0.06*					*+0.09*
L-O3	NOx	O_3	CH_4	CO		NOx	O_3	CH_4	CO
NOx	*+0.03*	**++0.05**	**++0.01**	**--0.01**		*0.02*	**++0.04**	**++0.01**	**--0.01**
O_3		*+0.23*	**++0.05**	**++0.02**			*+0.28*	**++0.04**	**--0.02**
CH_4			*-0.03*	**++0.02**				*-0.03*	**++0.03**
CO				*0.00*					*0.00*
L-CH4	NOx	O_3	CH_4	CO		NOx	O_3	CH_4	CO
NOx	*+0.09*	**--0.13**	**--0.18**	**++0.12**		*+0.08*	**--0.12**	**--0.18**	**++0.14**
O_3		*+0.39*	**--0.19**	**--0.01**			*+0.31*	**--0.17**	**--0.01**
CH_4			*+0.69*	**--0.12**				*+0.73*	**--0.10**
CO				*-0.34*					*-0.40*