Supplement of Atmos. Chem. Phys., 21, 8915–8932, 2021
https://doi.org/10.5194/acp-21-8915-2021-supplement

Supplement of

Radiative and chemical implications of the size and composition of aerosol particles in the existing or modified global stratosphere

Daniel M. Murphy et al.

Correspondence to: Daniel M. Murphy (daniel.m.murphy@noaa.gov)

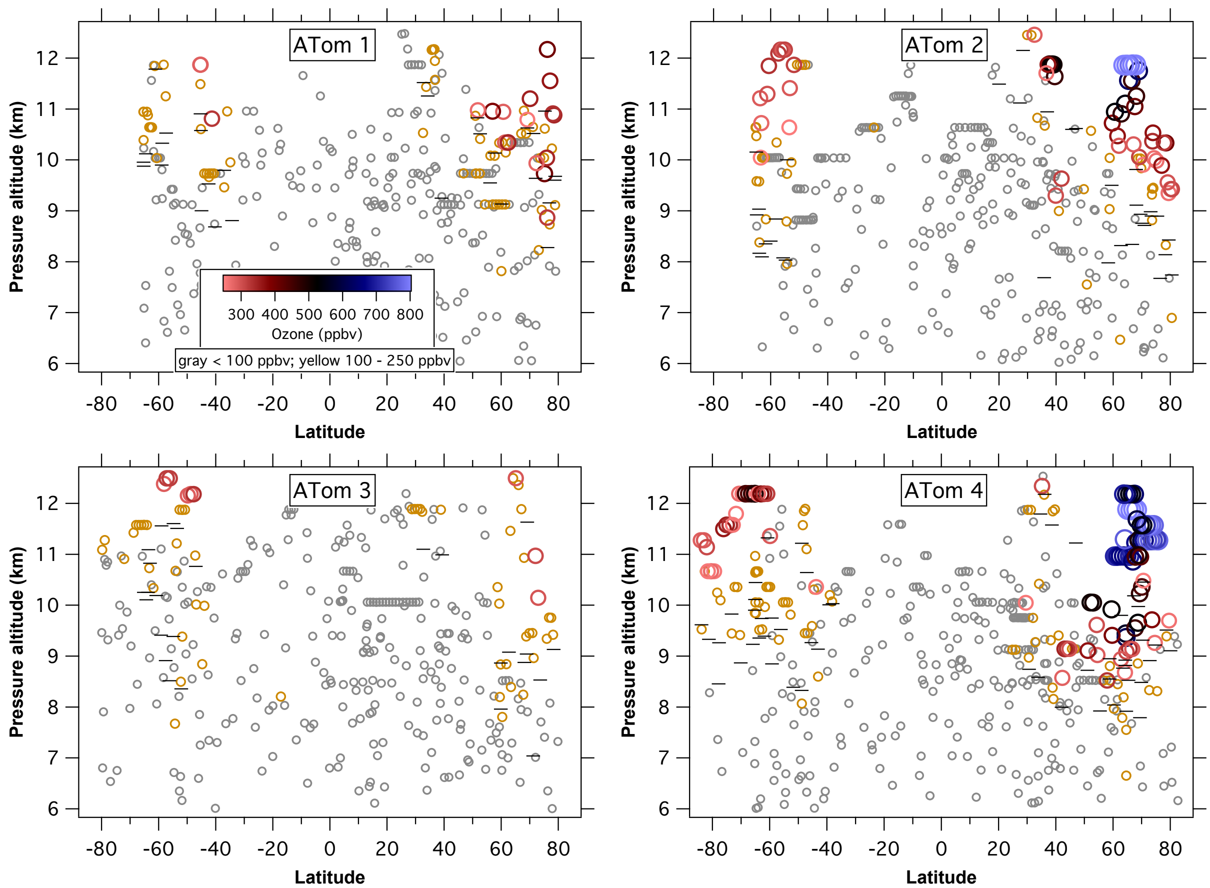

Figure S1. Locations of air with more than 100 or 250 ppbv ozone sampled during the ATom deployments. Each point represents 500 seconds of cloud-free air when PALMS was sampling. Small horizontal bars indicate locations of the lapse rate thermal tropopause when the DC8 crossed it during an ascent or descent. Other tropopause crossings were in level flight, where the thermal tropopause is hard to diagnose, and are not shown.

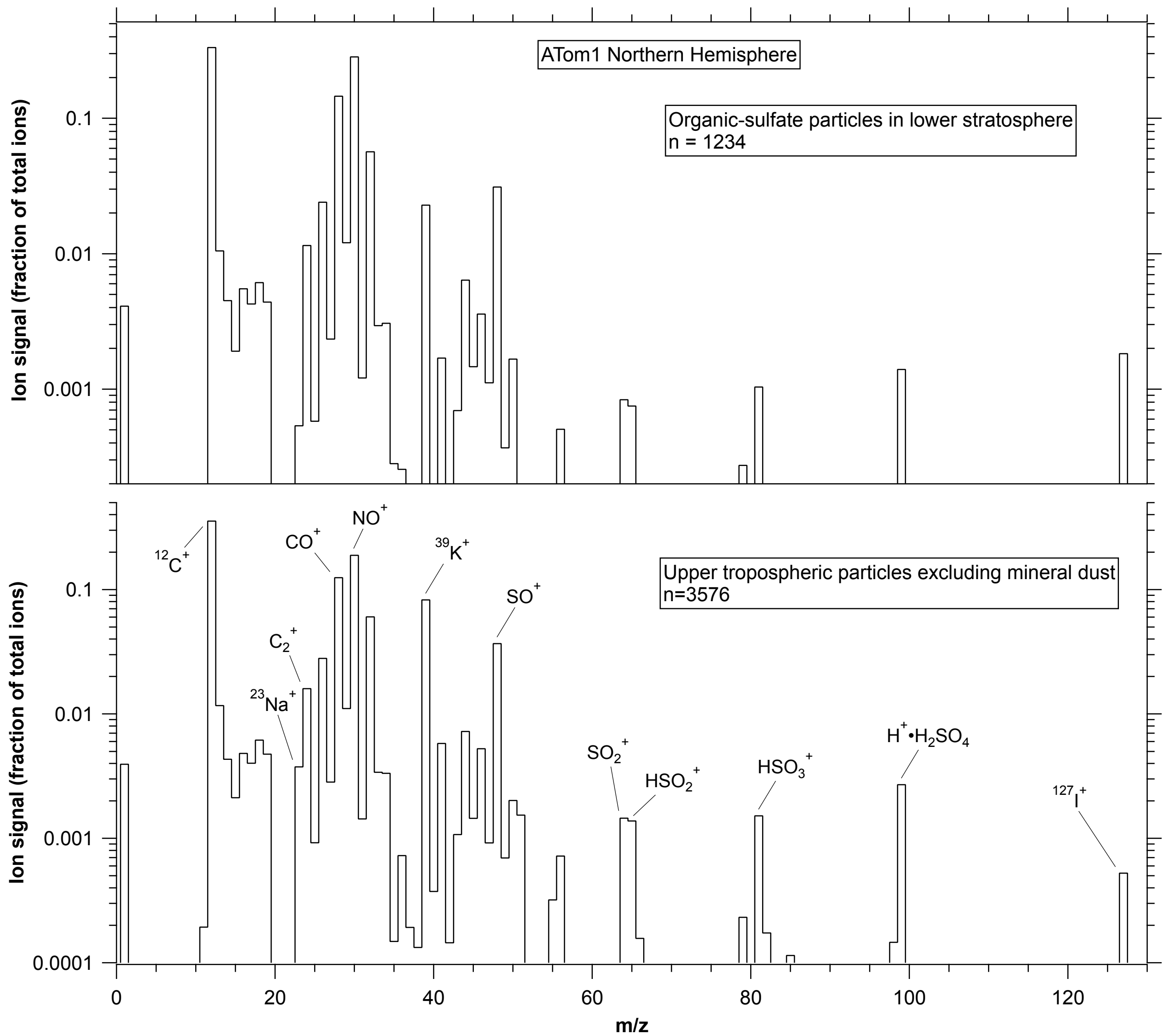

Figure S2. A comparison of the average positive ion mass spectra of organic-sulfate particles smaller than 0.35 µm in the lower stratosphere (ozone > 250 ppbv) with that of particles of similar size in the upper troposphere. Some major peaks with fairly unique identifications are labeled. Other deployments and locations showed similar results. The spectra strongly indicate that the organic-sulfate particles in the lower stratosphere originated in the troposphere.

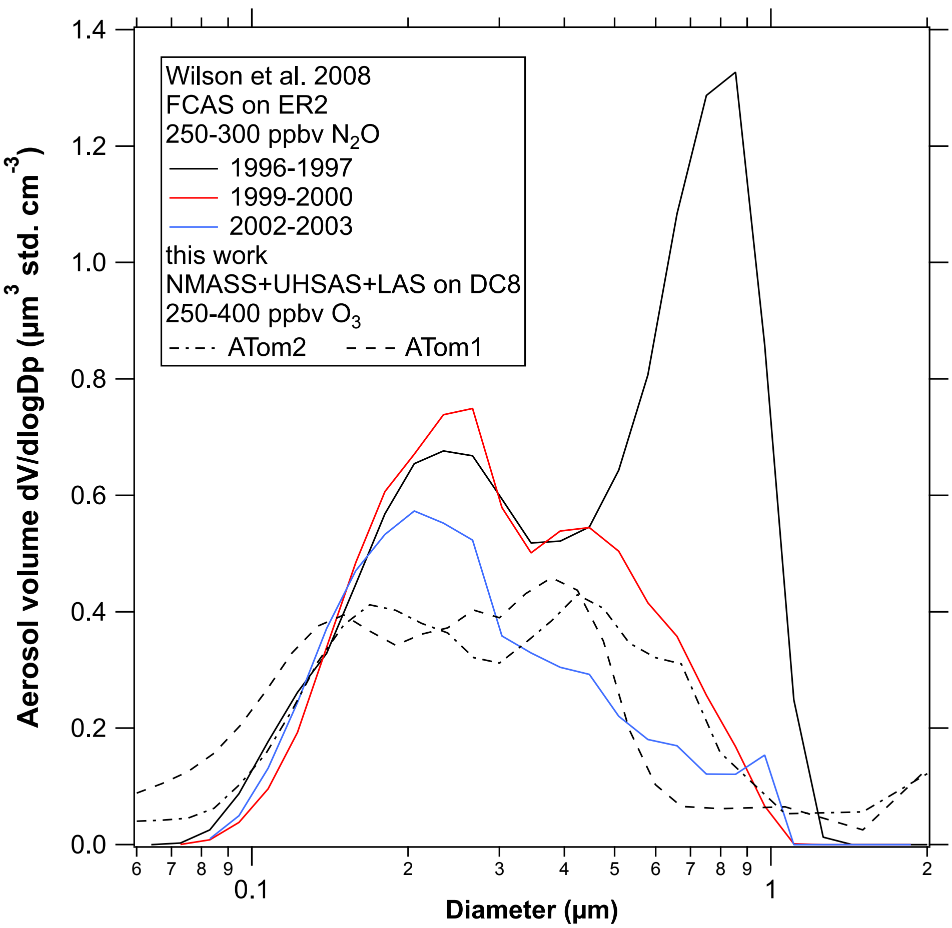

Figure S3. A comparison of this work with size distributions in Wilson et al. (2008). These are absolute concentrations instead of the normalized concentrations in the bottom panel of Figure 2 in Wilson et al. (2008). All data are for the Northern Hemisphere. 250 to 400 ppbv of ozone corresponds to roughly 275 to 300 ppbv of N_2O in the late 1990s. The 1996 Wilson et al. data show the aerosol still influenced by the Mt. Pinatubo eruption after the largest particles had sedimented out.

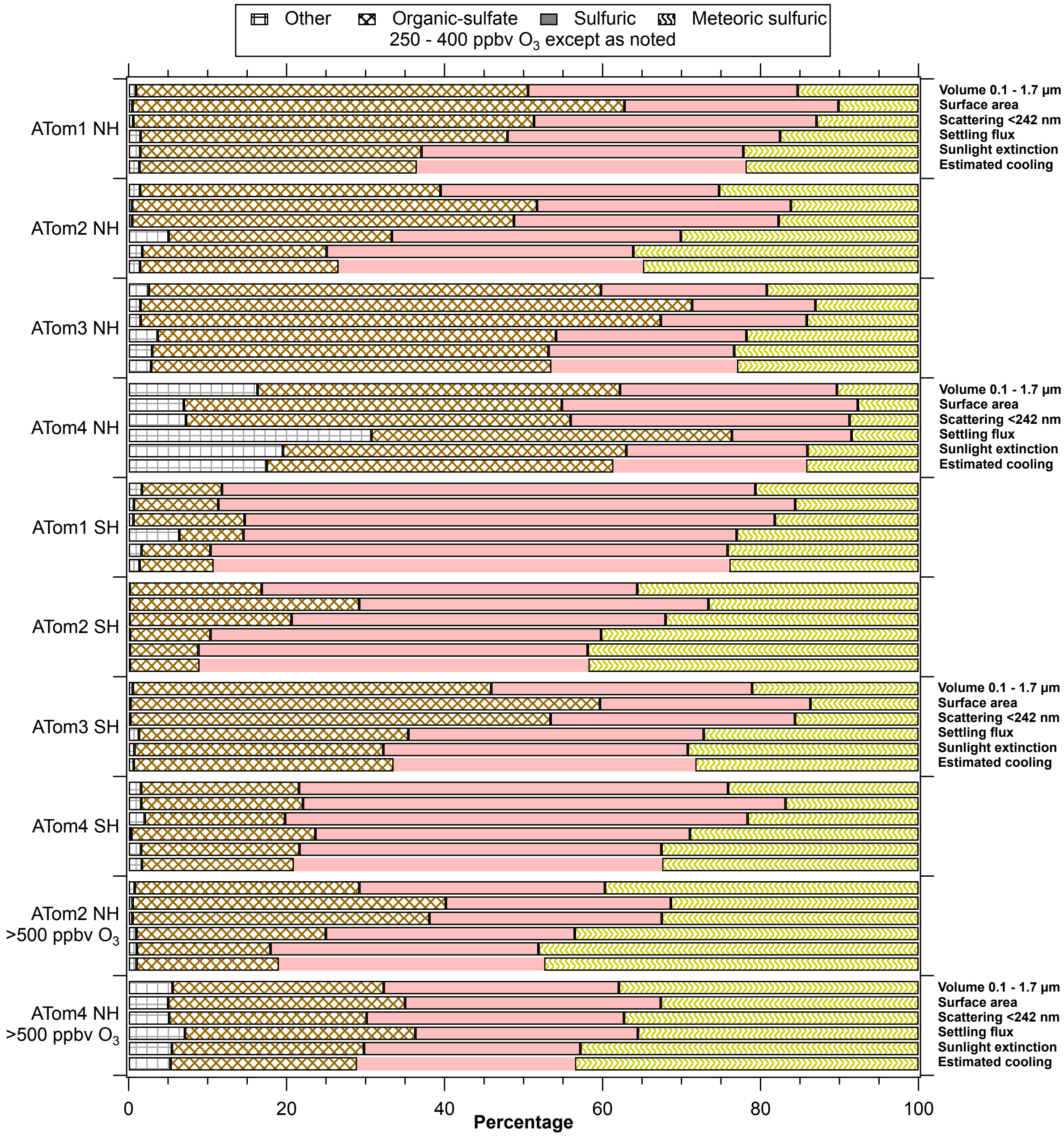

Figure S4. Percentages of various effects of dry aerosols in the lowermost stratosphere by type of particle. All calculations are for 0.1 to 1.7 µm diameter. Fractional abundances were extended to 0.1 µm when the data ended slightly above that (see Figure 1). Scattering <242 nm means scattering in the 210-242 nm range important for O_2 photolysis. Settling flux is for 20 km altitude pressure. The settling percentages are not very sensitive to pressure. Estimated cooling is backscatter of sunlight to outer space minus an offset independent of size to estimate the warming due to infrared absorption. Infrared heating for sulfuric acid was used for all particle types because there are insufficient infrared spectral data for the other particle types.

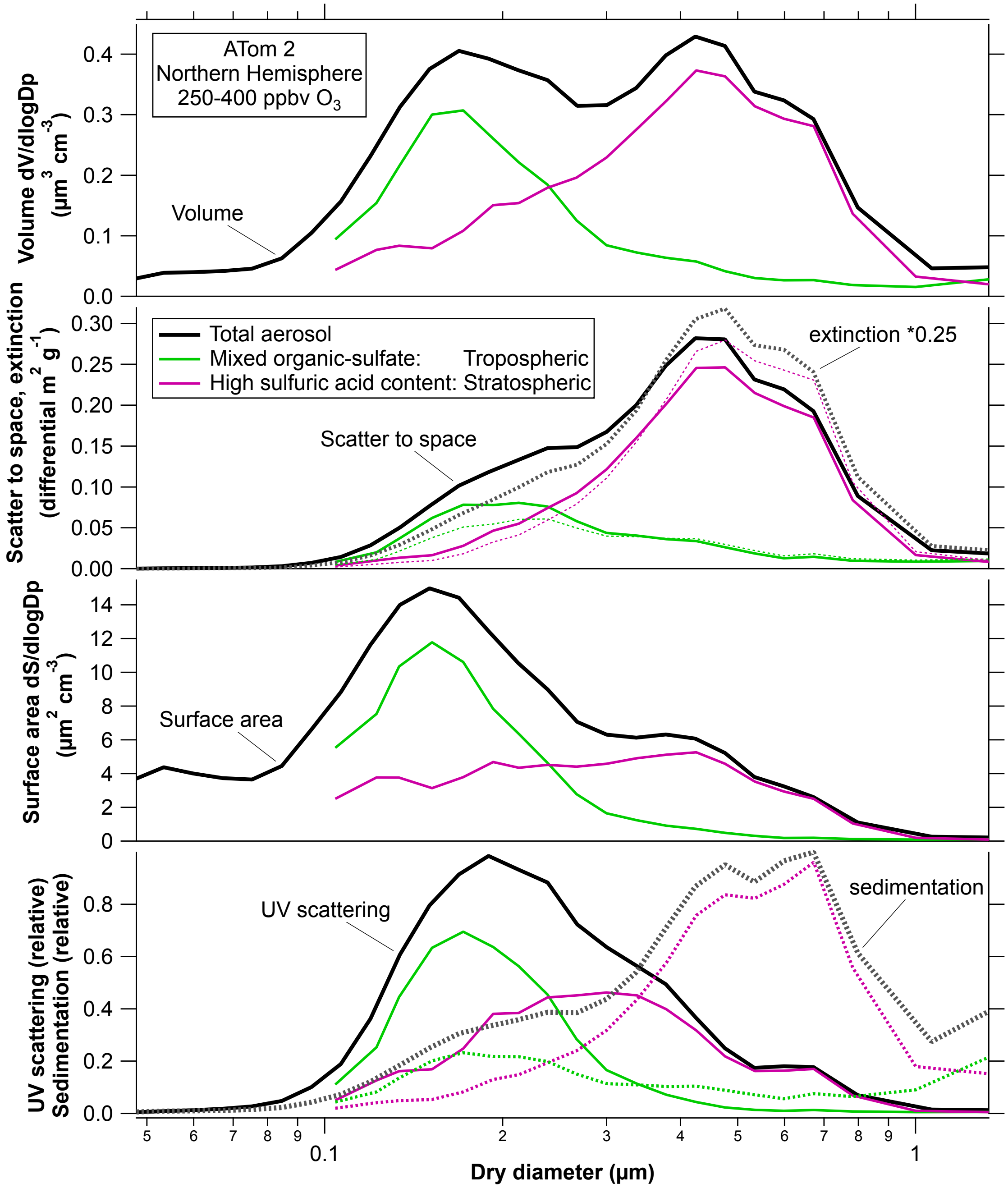

Figure S5. Similar to Figure 8 in the paper, data from ATom2 in the Northern Hemisphere. Size distributions weighted by extinction, UV scattering, and sedimentation are shown in addition to the volume, scatter to space, and surface area shown in Figure 8.

250 to 400 ppbv ozone						
Date	Latitude range		Longitude range		# positive	# negative
20160801	52.5	77.1	148.3 W	123.0 W	2416	2531
20160808	-45.5	-36.1	178.3 W	178.2E	1462	1168
20160812	-63.9	-57.5	145.1 W	79.5 W	149	89
20160820	69.2	78.8	77.0 W	49.7 W	2139	1816
20160822	51.7	61.0	85.8 W	70.4 W	957	850
20170126	31.4	33.3	121.0 W	119.7 W	380	240
20170129	49. 9	69.7	155.2 W	140.7 W	1607	1140
20170205	-54.0	-51.1	165.8 E	168.9 E	842	613
20170210	-65.3	-54.4	149.8 W	72.1 W	3021	1606
20170218	70.2	75.4	63.7 W	51.6 W	1793	1138
20170219	66.9	80.5	148.1 W	69.3 W	5920	3828
20170221	36.4	52.6	142.0 W	121.8 W	1584	1020
20171008	-52.8	-47.2	166.3 E	168.5 E	1707	1291
20171014	-63.9	-55.2	69.3 W	63.7 W	70	45
20171025	63.0	78.3	151.0 W	77.3 W	2580	1757
20180427	42.1	70.3	154.3 W	132.6 W	3498	2701
20180506	-65.1	-65.0	114.0 W	112.1 W	58	177
20180509	-86.0	-56.4	69.0 W	40.7 W	6336	3843
20180512	-44.2	-43.4	49.8 W	48.5 W	517	275
20180514	29.2	35.7	27.3 W	27.0 W	779	439
20180517	61.6	68.9	57.6 W	28.5 W	1596	856
20180519	57.5	80.0	148.9 W	80.5 W	4705	3544
20180521	38.3	60.2	148.3 W	121.9 W	1142	886
> 400 ppbv ozone						
20160801	61.8	75.9	134.0 W	125.5 W	531	417
20160820	69.9	78.5	75.8 W	54.1 W	341	201
20160822	51.8	56.7	85.7 W	78.8 W	227	99
20170129	59.7	69.5	155.0 W	146.6 W	6152	3619
20170219	67.6	78.9	148.1 W	112.4 W	1320	790
20170221	37.3	39.8	131.7 W	124.0 W	1027	676
20180427	54.5	70.2	154.1 W	142.4 W	7531	3901
20180509	-76.0	-62.4	68.5 W	63.3 W	641	416
20180517	64.4	75.9	66.9 W	42.6 W	4892	3173
20180519	57.3	73.9	148.9 W	80.2 W	7686	4503
20180521	51.0	60.1	148.0 W	141.0 W	2618	1630

Table S1. The number of positive and negative ion mass spectra acquired for two ranges of ozone characteristic of the stratosphere, along with the latitude and longitude of those spectra. Data are above 7 km, out of cloud, and for particles with an aerodynamic diameter acquired so they can be mapped to the size distributions. There are more positive ion spectra because PALMS was programmed to spend more time in positive ion mode. For reference, the total number of positive mass spectra above 7 km (both upper troposphere and stratosphere) was ATom1: 174451; ATom2: 160754; ATom3: 180033; ATom4: 294232.