Ambient aerosol properties in the remote atmosphere from global-scale in-situ measurements

Supplemental Materials

Four-mode lognormal fits to measured size distributions

Lognormal functions (Eq. 1) provide a concise description of the aerosol size distribution. Objectively fitting lognormal curves to multiple, dynamically varying modes spanning the diameter range from 3 nm to 50 µm is quite challenging. We have developed the following approach, which results in four lognormal functions that summed together accurately represent the number, surface, and volume of the entire size distribution. The coarse mode is first fitted using the portion of the volume size distribution with diameters from 1-6 µm, to avoid fitting noise from larger particles with poor counting statistics. The fitted mode diameter $D_{g,v}$ is constrained to lie between 1 and 10 µm, and the geometric standard deviation σ_g is constrained between 1.5 and 4. If the least-squares fit to Eq. 1 successfully converges, the volume median diameter is converted to number median diameter using the Hatch-Choate relationships (Heintzenberg, 1994) and the integrated volume converted to integrated number. This fitted size coarse-mode lognormal function is then subtracted from the original size distribution. Negative residual values are set to zero. The volume-weighted residual size distribution is then converted to volume space, and a lognormal fit is made to particles with diameters from 0.08-1 µm, with constraints of $D_{g,v}$ from 0.1-1.2 µm and of σ_g from 1.2-2.5. As before, the fitted distribution is converted to number space and subtracted from the residual size distribution. The Aitken and nucleation modes are subsequently each fitted directly to the residual number size distribution with constraints of $D_{g,n}$ from 0.01-0.2 µm and of σ_g from 1.2-2.5 over the size range from 0.008-0.2 µm for the Aitken mode, and constraints of $D_{g,n}$ from 0.002-0.01 µm and of σ_g from 1.2-2.5 over the size range from 0.003 to 0.012 µm for the nucleation mode.

A reconstructed size distribution is created by summing the four lognormal functions from the four modes. The validity of the fits is checked by integrating the number, surface, and volume of this reconstructed size distribution and comparing it to the measured size distribution over different size ranges. Least-squares regressions of these integrated values from the lognormal fits and the measured size distributions are provided in Tables S2-4. Slopes lie between 0.94 and 1.07 with r^2 values >0.76.

Table S1. Latitude boundaries for the regional air mass definitions for each ATom deployment.

Deployment	Arctic/midlatitude	North midlatitude/tropics	South midlatitude/tropics	Antarctic/midlatitude
ATom-1	60 °N	34 °N	21 °S	60 °S
ATom-2	60 °N	27 °N	32 °S	60 °S
ATom-3	61°N	29 °N	25 °S	60 °S
ATom-4	59 °N	20 °N	23 °S	60 °S

Table S2. Parameters from linear regression between integrated number concentration calculated from size distributions from four-mode lognormal fits to the measurements (ordinate) and integrated number directly from measurements (abscissa). Regression coefficients are from orthogonal distance (two-sided) regression; Pearson's regression coefficient (r^2) is from a one-sided fit. N is the number of 60s data points in the comparison.

Mode	Y-Intercept	Slope	r^2	N
Nucleation	-136 ± 5	0.979 ± 0.001	0.974	18742
Aitken	-93 ± 5	0.996 ± 0.003	0.761	18761
Accumulation	-9.9 ± 0.7	1.072 ± 0.002	0.939	18689
Coarse	0.036 ± 0.008	0.958 ± 0.002	0.905	18665

Table S3. As in Table S2, but for surface area.

Mode	Y-Intercept	Slope	r^2	N
Nucleation	-0.023 ± 0.001	0.977 ± 0.002	0.907	18742
Aitken	-0.29 ±-.01	0.990 ± 0.002	0.903	18761
Accumulation	-0.20 ± 0.04	1.037 ± 0.002	0.953	18689
Coarse	0.04 ± 0.09	0.991 ± 0.004	0.783	18665

Table S4. As in Table S2, but for volume.

Mode	Y-Intercept	Slope	r^2	N
Nucleation	1.8e-5 ± 2E-6	0.943 ± 0.003	0.836	18742
Aitken	-0.002 ± 0.0001	0.988 ± 0.002	0.920	18761
Accumulation	-0.0054± 0.002	1.030 ± 0.002	0.945	18689
Coarse	0.12± 0.03	1.022 ± 0.0)3	0.872	18665

Table S5. BC mass mixing ratio (MMR; ng g^{-1}_{air}), number geometric mean diameter (D_g, nm) for BC core assuming mass-equivalent sphere, geometric standard deviation σ_g, thickness of non-absorbing coating (CT, nm) and calculated mass absorption cross section (MAC, $m^2 g^{-1}$) at 532 nm for the different regions and air mass types. Data from the northern and southern hemisphere stratosphere have been combined.

Free Troposphere	**ATom-1**					**ATom-2**					**ATom-3**					**ATom-4**				
Pacific	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC
Antarctic/S.Ocean	1.7	108	1.5	78	14.6	0.6	96	1.52	102	17.2	8.3	110	1.5	76	14.3	0.3	84	1.57	60	14.4
S. Midlatitudes	2	105	1.51	75	14.5	1.3	73	1.66	76	15.9	12.1	107	1.51	70	13.9	2	79	1.65	54	13.3
Tropics	2	91	1.58	58	13.3	2.6	94	1.6	81	14.6	4	96	1.53	61	13.9	0.9	76	1.63	64	14.8
N. Midlatitudes	9.9	83	1.62	72	14.9	9.3	97	1.63	102	14.9	9.3	84	1.65	71	14.2	18.8	84	1.6	65	14.4
Arctic	4.4	58	1.72	60	15.5	7.1	85	1.73	84	14.1	7.4	81	1.64	114	17.3	10.4	92	1.58	72	14.5
Atlantic																				
Antarctic/S.Ocean	–	–	–	–	–	–	–	–	–	–	2.4	115	1.49	77	14.0	0.3	87	1.63	66	13.7
S. Midlatitudes	3.5	106	1.52	67	13.6	0.2	102	1.52	85	15.4	7.9	98	1.58	61	12.9	0.9	93	1.54	51	13.1
Tropics	28.2	134	1.44	84	13.6	12.5	123	1.49	67	12.4	19.5	95	1.56	47	12.2	2.9	105	1.51	44	11.8
N. Midlatitudes	2.9	51	1.8	59	15.7	3.1	77	1.7	71	14.5	4.9	66	1.75	64	14.7	12.4	95	1.56	71	14.4
Arctic	2.7	63	1.74	83	16.9	4.3	75	1.7	73	14.9	7.7	62	1.82	75	15.8	11.8	98	1.53	76	15.0
Marine Boundary Layer																				
Pacific	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC
Antarctic/S.Ocean	0.1	93[A]	1.6[A]	60[A]	13.0	0.1	93[A]	1.6[A]	60[A]	13.0	0.2	93[A]	1.6[A]	60[A]	13.0	0	93[A]	1.6[A]	60[A]	13.0
S. Midlatitudes	0.3	96	1.57	62	13.4	2.5	44	1.87	53	15.6	7.1	108	1.52	67	13.4	21.9	94	1.54	74	15.1
Tropics	13.1	98	1.53	58	13.4	3.5	106	1.54	84	14.6	9.6	112	1.48	65	13.6	17.6	98	1.54	61	13.5
N. Midlatitudes	1	93[A]	1.6[A]	60[A]	13.0	5.9	104	1.62	119	14.6	2.7	84	1.67	77	14.4	18.8	97	1.55	73	14.5
Arctic	0.5	93[A]	1.6[A]	60[A]	13.0	48.6	132	1.62	91	10.9	0.7	93[A]	1.6[A]	60[A]	13.0	6.7	95	1.6	60	12.7
Atlantic																				
Antarctic/S.Ocean	–	–	–	–	–	–	–	–	–	–	1.2	119	1.48	74	13.6		93[A]	1.6[A]	60[A]	13.0
S. Midlatitudes	42.7	122	1.47	57	12.1	0.3	93	1.6	52	12.3	20.7	90	1.61	44	11.7	0.5	93	1.6	57	12.7
Tropics	581.2	135	1.44	77	13.0	77.4	126	1.47	65	12.4	91.9	91	1.61	48	12.0	25	101	1.52	44	12.0
N. Midlatitudes	10.8	61	1.76	57	14.6	3	85	1.68	75	14.0	4.9	77	1.69	78	15.2	7.9	91	1.57	60	13.7
Arctic	0.7	38	2.06	101	20.8	7.9	107	1.54	81	14.2	0.4	93[A]	1.6[A]	60[A]	13.0	3.3	106	1.5	71	14.3
	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC	MMR	D_g	σ_g	CT	MAC
Stratosphere	2	73	1.64	63	14.4	0.9	59	1.65	71	16.6	9.1	108	1.5	86	14.7	2.5	77	1.62	144	17.8
Biomass Burning Plumes	164.7	129	1.46	85	13.3	71.2	138	1.41	75	12.7	31.3	102	1.52	70	13.8	37.1	103	1.52	67	13.5
Dust Plumes	37.9	90	1.62	75	13.8	164.6	145	1.39	67	11.9	80.5	92	1.59	47	11.8	9.1	91	1.61	67	13.2

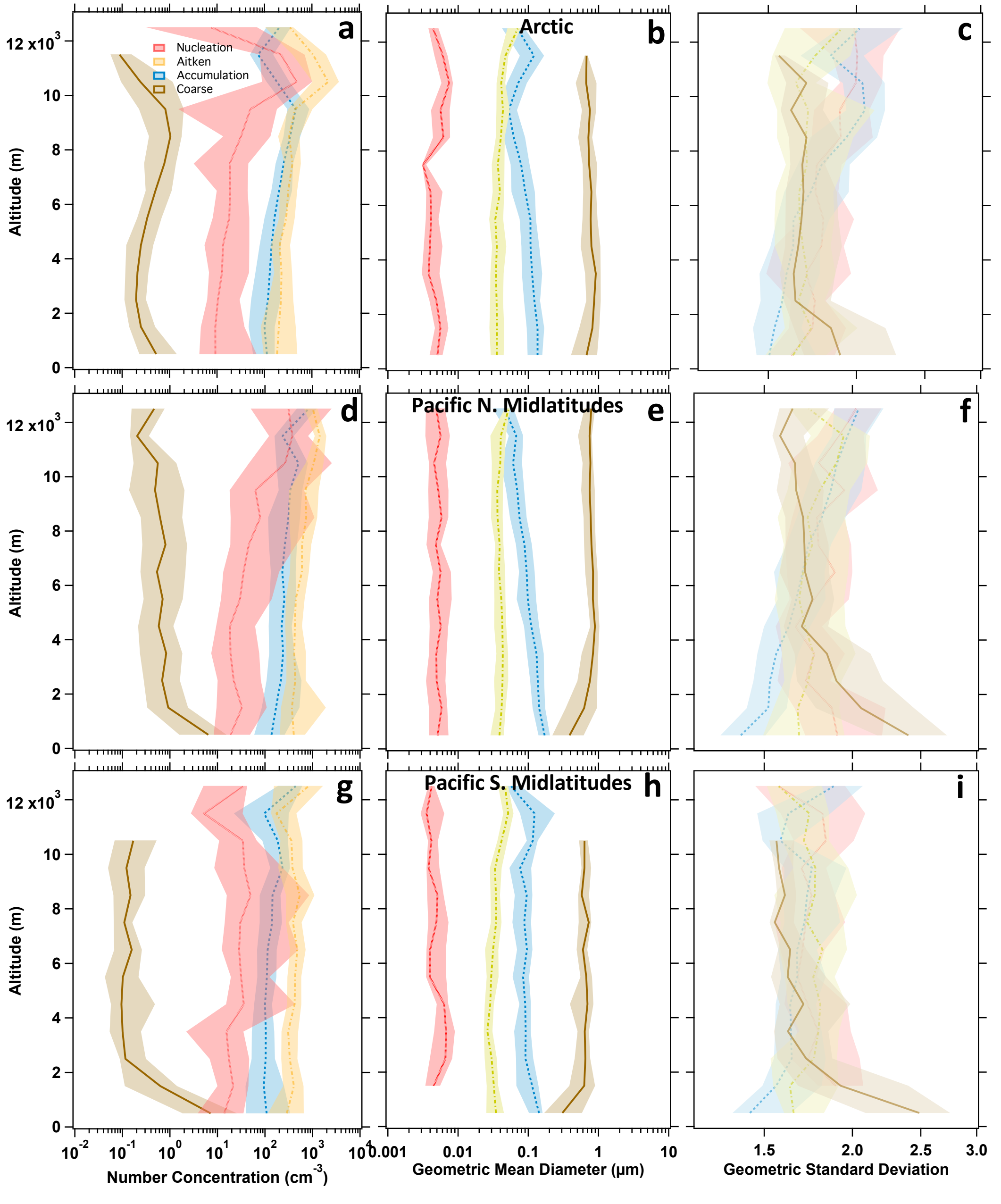
[A]Insufficient data for statistics; values are assumed.

Table S6. Lognormal parameters geometric standard deviation σ_g, number geometric mean diameter $D_{g,n}$, and volume geometric mean diameter $D_{g,v}$ for aerosol types from the OPAC database (Hess et al., 1998), from global shipborne data (Quinn et al., 2017), from the 7-mode modal aerosol model (MAM7; Liu et al., 2012), and from the ATom measurements. Atom values encompass the 25th to 75th percentiles; FT data are between 2 and 12 km. Quinn et al. data represent the full range. MAM7 ranges show the 10th and 90th percentiles. A subset of these data is shown in Fig. 15.

Component	σ_g	$D_{g,n}$ (μm)	$D_{g,v}$ (μm)
OPAC water-soluble	2.24	0.042	0.30
ATom N. Pac. FT accumulation mode	1.44-2.09	0.08-0.11	0.18-0.30
ATom BB accumulation mode	1.54-1.78	0.08-0.13	0.20-0.25
MAM7 accumulation mode	1.8	0.056-0.26	—
ATom N. Pac. FT Aitken mode	1.55-2.12	0.029-0.052	0.068-0.20
ATom MBL Aitken mode	1.57-1.89	0.029-0.047	0.065-0.14
Quinn et al. MBL Aitken mode	1.3-1.8	0.01-0.08	—
MAM7 Aitken mode	1.6	0.015-0.052	—
OPAC soot	2.00	0.022	0.10
ATom free troposphere BC[1]	1.49-1.82	0.14-0.21	0.071[2]
ATom biomass burning BC[1]	1.41-1.52	0.17-0.20	0.074[2]
MAM7 primary carbon	1.6	0.039-0.13	—
OPAC sea-salt accumulation mode[3]	2.03	0.42	1.9
ATom MBL accumulation mode[3]	1.32-1.53	0.13-0.18	0.21-0.25
Quinn et al. MBL accumulation mode	1.3-1.8	0.09-0.23	—
MAM7 fine sea-salt mode	2.0	0.095-0.56	—
OPAC sea-salt coarse mode	2.03	3.50	15.8
ATom MBL coarse mode	2.09-3.13	0.27-0.97	2.9-5.1
Quinn et al. MBL sea spray mode	2.2-2.8	0.17-0.45	—
MAM7 coarse sea-salt mode	2.0	0.63-3.7	—
OPAC mineral accumulation mode[4]	2.00	0.78	3.20
ATom dust accumulation mode[4]	1.63-2.04	0.07-0.14	0.24-0.40
MAM7 fine dust mode	1.8	0.14-0.62	
OPAC mineral coarse mode	2.15	3.80	22.0

OPAC mineral-transported mode[5]	2.20	1.00	6.00
ATom dust coarse mode	1.77-2.01	0.88-1.09	2.7-4.1
MAM7 coarse dust mode	1.8	0.59-2.75	—

[1]Parameters for mass-equivalent sphere of uncoated BC
[2]Mean thickness of coating on BC
[3]A separate accumulation mode of sea-salt particles is not observed in the ATom dataset, but coarse-mode sea-salt particles extend into the accumulation mode. ATom modal parameters in the table are for the observed sulfate/organic-dominated accumulation mode.
[4]A separate accumulation mode of mineral particles is not observed in the ATom dataset. ATom modal parameters in the table are for the observed sulfate/organic-dominated accumulation mode.
[5]As described in Hess et al. (1998), "mineral-transported" refers to "desert dust that is transported over long distances with a reduced amount of large particles."



65 **Figure S1. Vertical profiles of fitted lognormal parameters for the nucleation, Aitken, accumulation and coarse modes for the Arctic (a, b, and c), Pacific northern midlatitudes (d, e, and f), and the Pacific southern midlatitudes (g, h, and i) for the entire ATom project. Lines are median values and shaded regions show the interquartile range. Similar vertical profiles for the tropics of the Pacific and Atlantic are in Fig. 11 of the main text.**

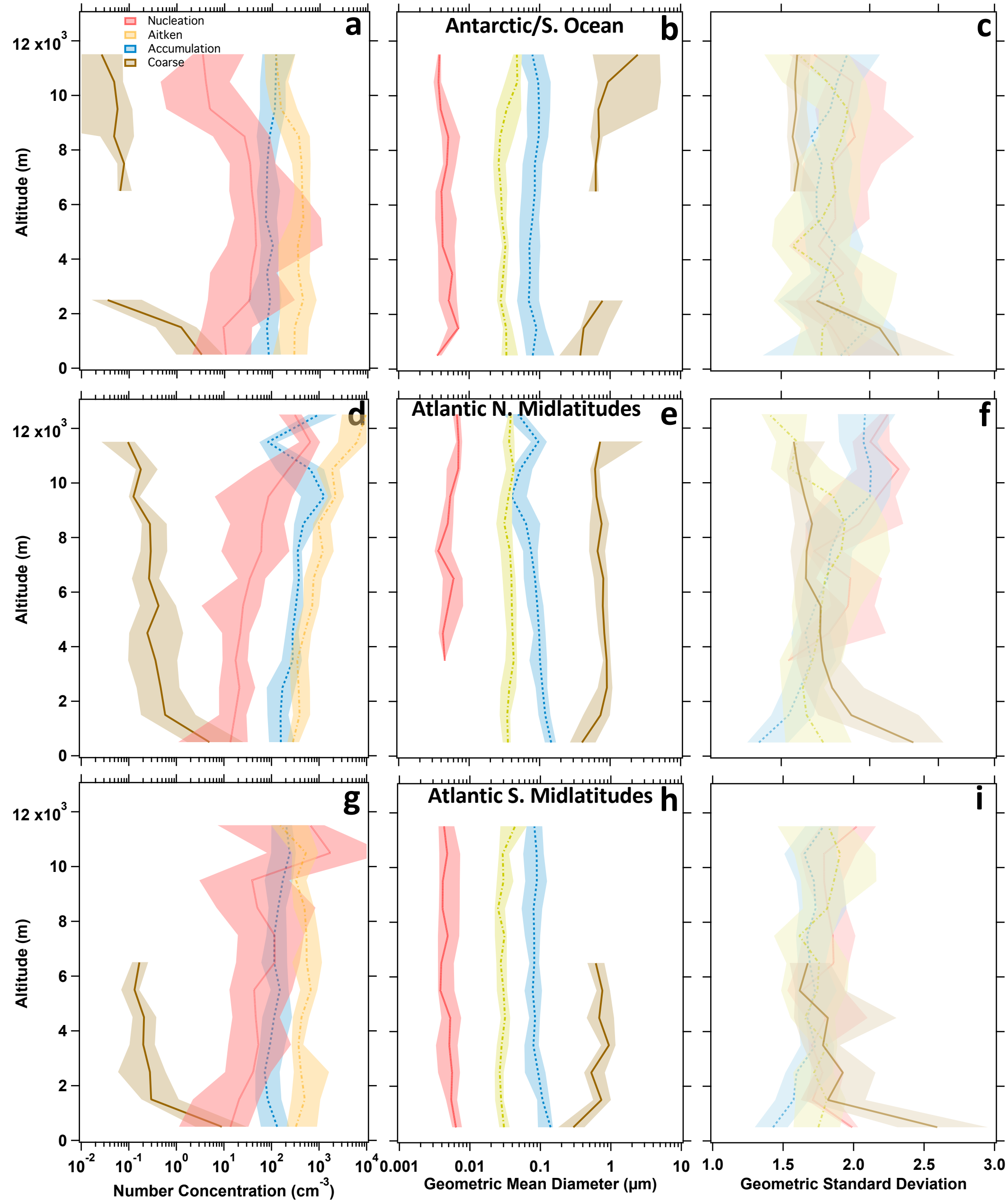


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Figure S2. Vertical profiles of fitted lognormal parameters for the nucleation, Aitken, accumulation and coarse modes for the Antarctic and Southern Ocean (a, b, and c), Atlantic northern midlatitudes (d, e, and f), and the Atlantic southern midlatitudes (g, h, and i) for the entire ATom project. Lines are median values and shaded regions show the interquartile range. Similar vertical profiles for the tropics of the Pacific and Atlantic are in Fig. 12 of the main text.

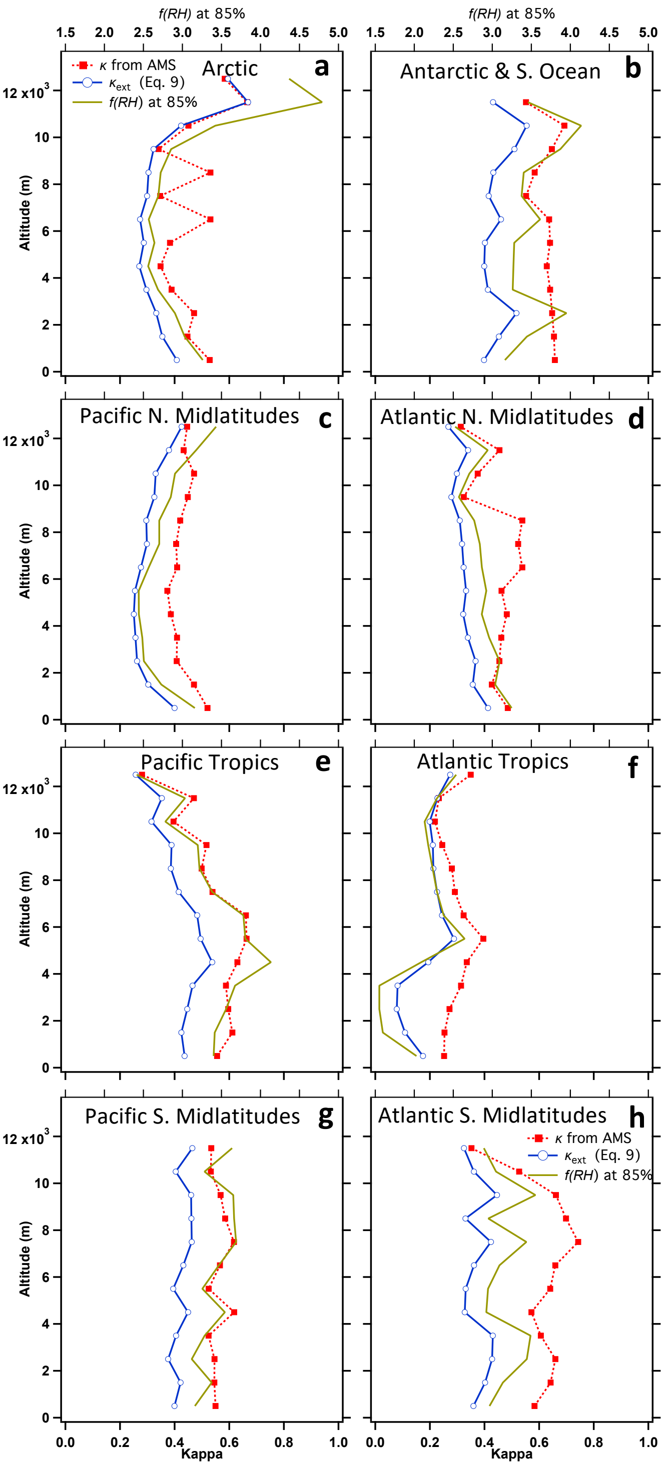

Figure S3. Median vertical profiles of κ determined from the AMS composition as described in Sect. 2.5 (bottom axis), of optical κ_{ext} values from Eq. 9 (bottom axis) and of *f(RH)* as described in Sect. 2.7.2 (top axis) for different regions sampled during the entire ATom project. a) Arctic. b) Antarctic and Southern Ocean. c) Pacific northern midlatitudes. d) Atlantic northern midlatitudes. e) Pacific tropics. f) Atlantic tropics. g) Pacific southern midlatitudes. h) Atlantic southern midlatitudes

References

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