S1. GRASP-PFR validation

Table S1: Statistics of the differences between GRASP-PFR retrievals and AER-SKY for AOD at 500 nm > 0.1. The properties corresponding to fine mode correspond also to AOD_f>0.04 and AE>1, while the coarse mode properties, to AOD_c>0.04 and AE<1.5. We also include the AOD at 500 nm comparison between the PFR and AER-SKY.

Parameter	**mean**	**median**	**St.d.**	**P95-P5**	**R**	**median of the parameter - reference**	**Number of measurements**
AOD 500 nm	-0.003	-0.003	0.009	0.027	0.99	0.154	1500
AOD_f 500 nm	-0.002	-0.002	0.013	0.040	0.98	0.131	1170
AOD_c 500 nm	-0.003	-0.003	0.007	0.022	0.99	0.114	356
C_{VT}	-0.008	-0.006	0.015	0.047	0.94	0.047	1500
C_{Vf}	-0.001	-0.002	0.007	0.022	0.79	0.021	1170
C_{Vc}	-0.022	-0.019	0.018	0.057	0.93	0.082	356
R_{eff}	-0.084	-0.004	0.225	0.722	0.91	0.306	1500
R_{Vf}	0.018	0.019	0.023	0.075	0.71	0.174	1170
R_{Vc}	-0.368	-0.304	0.337	1.109	0.10	1.750	356

Median uncertainties:

AER-SDA:

U-AOD_f=0.010

U-AOD_c=0.006

AER-SKY:

U-R_{Vf}=0.01 µm

U-R_{Vc}=0.096 µm

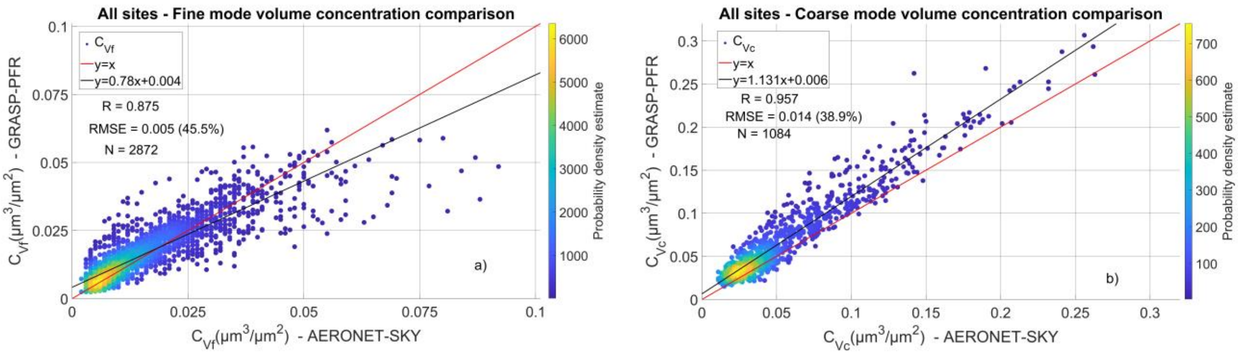

Figure S1: Scatter plot of C_{Vf} (a) and C_{Vc} (b) for the GRASP-PFR and AER-SKY retrievals from all four locations. The plots include the correlation factor (R), the root mean square error (RMSE) and the number of observations (N). The colour bar shows the density of the points. We also include the linear fit between the datasets and the y=x line.

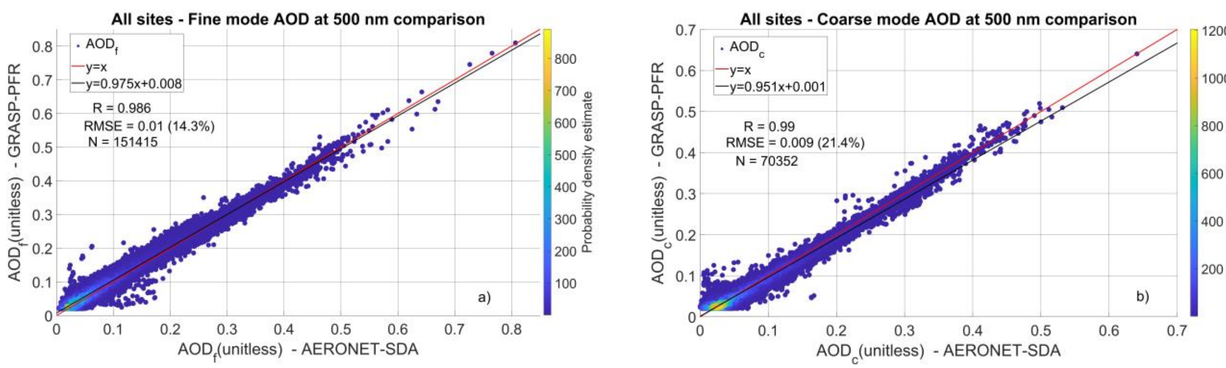

Figure S2: Scatter plot of AOD_f (a) and AOD_c (b) for the GRASP-PFR and AER-SKY retrievals from all four locations.

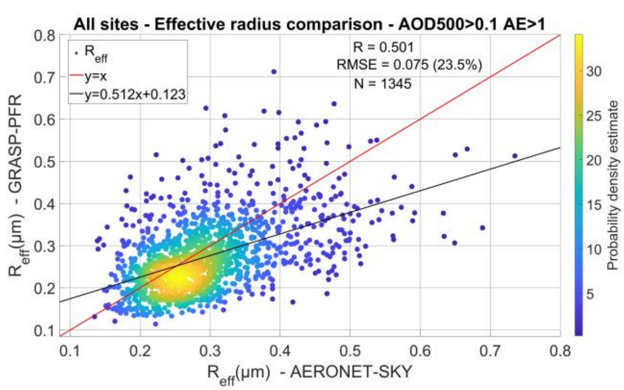

Figure S3: Scatter plot of R_{eff} for the GRASP-PFR and AER-SKY retrievals for data corresponding to AOD>0.1 at 500 nm and AE>1.

S2. Data screening

AERONET inversions data screening:

We rejected all data corresponding to AOD, AOD_f and AODc differences at 500 nm between AERONET inversions from almucantar scans and retrievals by the AERONET spectral deconvolution algorithm according to the following empirically defined thresholds of keeping data:

1. Coincident AOD, AOD_f and AOD_c differences at 500 nm <0.03 (7.5 minutes seconds threshold).
2. Common daily AOD median differences <0.015 for AOD and <0.018 for AOD_f and AOD_c.
3. 80th-20th percentile of daily AOD, AOD_f and AOD_c difference <0.055.

Direct sun AOD data screening:

We rejected data corresponding to AOD differences (30 seconds threshold) between PFR and CIMEL according to the following thresholds:

1. Coincident AOD differences <0.09 for 380 and 440 nm, <0.06 for 500 nm and <0.04 for 870 nm.
2. Common daily AOD median differences <0.05 for 380 and 440 nm, <0.014 for 500 nm and <0.009 for 870 nm.
3. 80th-20th percentile of daily AOD difference <0.09 for 380 and 440 nm, <0.03 for 500 nm and <0.02 for 870 nm.

S3. Refractive index climatologies

Table S2: Refractive index climatologies for all stations.

Month	**RRI**				**IRI**			
	440 nm	675 nm	870 nm	1020 nm	440 nm	675 nm	870 nm	1020 nm
Davos								
Annual for all months	1.378	1.441	1.465	1.4621	0.0073	0.0026	0.0021	0.0018
Hohenpeissenberg								
Annual for all months	1.469	1.459	1.453	1.448	0.0032	0.0035	0.0038	0.0039
Izana								
1	1.476	1.472	1.448	1.436	0.0023	0.0006	0.0009	0.0012
2	1.476	1.472	1.448	1.436	0.0023	0.0006	0.0009	0.0012
3	1.476	1.472	1.448	1.436	0.0023	0.0006	0.0009	0.0012
4	1.484	1.477	1.454	1.442	0.0024	0.0007	0.0012	0.0016
5	1.484	1.477	1.454	1.442	0.0024	0.0007	0.0012	0.0016
6	1.484	1.477	1.454	1.442	0.0024	0.0007	0.0012	0.0016
7	1.480	1.475	1.452	1.440	0.0023	0.0006	0.0010	0.0013
8	1.480	1.475	1.452	1.440	0.0023	0.0006	0.0010	0.0013

9	1.475	1.473	1.449	1.438	0.0023	0.0005	0.0008	0.0010
10	1.475	1.473	1.449	1.438	0.0023	0.0005	0.0008	0.0010
11	1.475	1.473	1.449	1.438	0.0023	0.0005	0.0008	0.0010
12	1.476	1.472	1.448	1.436	0.0023	0.0006	0.0009	0.0012
Lindenberg								
1	1.489	1.474	1.469	1.461	0.0070	0.0068	0.0083	0.0090
2	1.489	1.474	1.469	1.461	0.0070	0.0068	0.0083	0.0090
3	1.469	1.462	1.461	1.456	0.0072	0.0070	0.0081	0.0086
4	1.469	1.462	1.461	1.456	0.0072	0.0070	0.0081	0.0086
5	1.453	1.453	1.453	1.450	0.0061	0.0057	0.0066	0.0069
6	1.471	1.461	1.457	1.450	0.0050	0.0048	0.0057	0.0060
7	1.457	1.451	1.449	1.444	0.0054	0.0056	0.0067	0.0071
8	1.461	1.453	1.451	1.445	0.0055	0.0059	0.0069	0.0073
9	1.449	1.446	1.446	1.442	0.0057	0.0063	0.0073	0.0077
10	1.470	1.461	1.459	1.453	0.0051	0.0053	0.0059	0.0060
11	1.470	1.461	1.459	1.453	0.0051	0.0053	0.0059	0.0060
12	1.479	1.467	1.464	1.457	0.0061	0.0061	0.0071	0.0075

S4. Scenarios, weights and rating for GRASP self-consistency tests

The settings are selected depending on the performance of self-consistency tests, AOD and AE. The self-consistency tests are evaluated through case-by-case inspection and a rating system. We selected 10 scenarios for the parameters C_{Vf}, C_{Vc}, R_{Vf} and R_{Vc} corresponding to various usual and unusual or even extreme conditions (Table S3). We also assigned for each scenario a weight based on the frequency it appears on the AERONET data included in the GRASP-PFR validation in the four common stations. For each scenario we calculate the AOD using the GRASP forward model and provide that AOD (Table S4) to the inversion mode of GRASP to retrieve back the aerosol size properties we selected. Then we calculate the difference of the aerosol size properties and AOD between the forward model and inversion to evaluate the self-consistency. The procedure is repeated for various initial guesses treating one parameter as variable every time. Each parameter includes a weight based on the performance in the PFR validation and an uncertainty based on either the GAW-PFR or AERONET uncertainty estimates (AOD) or the standard deviation of the differences between GRASP-PFR and AER-SKY (Table S4).

The weights of each scenario and parameter and the uncertainty estimates in combination with the differences between the forward model and inversion, provide a combined rating according to Eq. S1. The lower the rating, the more self-consistency the inversion showed.

After a provisional selection of initial guesses, we repeated the self-consistency tests for all their combinations to evaluate the performance and find potentially unstable combinations. From the combinations that showed no unusually worse self-consistency from the rest, we selected initial guesses based on the AOD and AE that they correspond according to the forward model (Table S8). The final selection is in the end of this section after table S8.

Provisional selection of initial guesses:

C_{Vf} and C_{Vc}: 0.001, 0.005, 0.006, 0.008, 0.009

R_{Vf}: 0.1, 0.15, 0.225

R_{Vc}: 1.75, 2.25

Table S3: Scenarios for forward model runs used for the self-consistency tests to retrieve aerosol properties from BTS spectroradiometer. Fixed parameters: RRI=1.49, IRI=0.009, σ_{Vf}=0.4 and σ_{Vc}=0.7.

Scenario number	**Scenario name**	**C_{Vf} ($\mu m^3/\mu m^2$)**	**C_{Vc} ($\mu m^3/\mu m^2$)**	**R_{Vf} (μm)**	**R_{Vc} (μm)**	**AOD 500nm**	**Weight_scen**
1	lrhc	0.1	0.12	0.4	4	0.722	0.01
2	srhc	0.1	0.12	0.1	1.5	0.626	0.01
3	lrlc	0.003	0.005	0.4	4	0.022	0.01
4	srlc	0.003	0.005	0.1	1.5	0.021	0.01
5	moder1	0.01	0.035	0.15	1.8	0.114	1
6	moder2	0.01	0.035	0.2	2.5	0.112	1
7	mrhc	0.1	0.12	0.15	2.5	0.398	0.02
8	mrlc	0.1	0.12	0.15	2.5	0.025	0.01
9	mrhflc	0.05	0.005	0.15	2.5	0.338	0.05
10	mrlfhc	0.003	0.07	0.15	2.5	0.085	0.15

Table S4: Weights and uncertainties (AOD parameters) or standard deviations of the differences in the PFR comparisons (aerosol size properties and residuals) for each parameter.

Parameter	**Weight_par**	**σ_{par}**
ΔC_{Vf}	0.85	0.006
ΔC_{Vc}	0.85	0.015
ΔR_{Vf}	0.6	0.038
ΔR_{Vc}	0.1	0.503
$\Delta \sigma_{Vf}$	0.01	0.168
$\Delta \sigma_{Vc}$	0.01	0.090
res. rel.	0.50	0.021
res. abs.	1	0.002
mean ΔAOD	0.75	0.010
median ΔAOD	1	0.010
ΔAOD st.d.	0.75	0.010

Rating calculation:

$$Rating = \sum_{scenn=1}^{10}(Weight\_scenn \times \sum_{parn=1}^{11}(|100 \times \frac{Weight_{par}*\Delta par}{\sigma_{par}}|)) \quad \text{(S1)}$$

Table S5: Initial guesses of C_{Vf} and C_{Vc} and the corresponding rating for each wavelength selection.

Initial guess	**Rating**		**Initial guess**	**Rating**	
C_{Vf}	16wl	7wl	C_{Vc}	16wl	7wl
0.001	16.9	1310.4	0.001	13.5	2759.5
0.003	33.8	2734.7	0.003	80.6	311.0
0.005	14.0	2803.9	0.005	12.1	273.1
0.006	82.8	2755.7	0.006	49.5	248.0
0.007	13.5	2759.5	0.007	95.2	266.3
0.008	12.5	2800.7	0.008	14.4	253.3
0.009	12.6	2852.0	0.009	12.4	263.2
0.01	13.3	2859.3	0.01	12.5	359.2
0.02	101.3	2877.2	0.03	25.2	354.9
0.03	137.4	2977.3	0.05	25.0	365.5
0.05	142.7	2860.2	0.09	13.4	393.0
0.09	218.0	2759.3	0.12	14.4	379.2

Table S6: Initial guesses of R_{Vf} and R_{Vc} and the corresponding rating for each wavelength selection.

Initial guess	**Rating**		**Initial guess**	**Rating**	
R_{Vf}	16wl	7wl	R_{Vc}	16wl	7wl
0.1	55.8	1476.6	1.75	3.5	308.6
0.125	14.6	3034.5	2	15.3	1549.4
0.15	12.5	2802.2	2.25	6.7	1680.3
0.175	106.7	2943.6	2.5	8.5	1759.2
0.2	130.3	3030.6	2.75	10.0	2688.2
0.225	25.6	2962.9	3	12.6	2734.2
0.25	2249.9	2948.8	3.25	57.4	2776.1
0.275	2338.7	2929.7	3.5	164.5	2812.9
0.3	4373.9	2915.8	3.75	353.4	2836.8
0.325	4486.1	2907.4	4	298.5	2865.9
0.35	4677.8	3273.2	-	-	-

0.375	4750.5	3115.5	-	-	-

Table S7: Initial guesses of IRI and the corresponding rating for each wavelength selection.

Input inv.	**Rating**	
IRI	16wl	7wl
0.001	80.3	2871.4
0.002	28.3	2857.1
0.003	25.7	2842.5
0.006	19.7	2797.5
0.009	13.5	2759.5
0.012	79.1	2724.1
0.015	29.4	2692.1
0.02	130.8	2682.5
0.04	93.3	2797.4
0.06	149.4	2868.5

Table S8: AOD and AE for different SD parameters from GRASP forward model runs.

C_{Vf} ($\mu m^3/\mu m^2$)	**C_{Vc} ($\mu m^3/\mu m^2$)**	**R_{Vf} (μm)**	**R_{Vc} (μm)**	**AOD at 500 nm**	**AE**
0.001	0.001	0.15	2.25	0.008	1.606
0.001	0.001	0.1	1.75	0.006	1.583
0.001	0.001	0.225	3	0.009	1.233
0.001	0.005	0.15	2.25	0.012	0.844
0.005	0.001	0.15	2.25	0.034	1.987
0.005	0.005	0.15	2.25	0.039	1.606
0.006	0.006	0.15	2.25	0.046	1.606
0.008	0.008	0.15	2.25	0.062	1.606
0.009	0.009	0.15	2.25	0.069	1.606
0.009	0.009	0.1	1.75	0.051	1.583
0.009	0.009	0.225	3	0.081	1.233
0.001	0.009	0.15	2.25	0.016	0.559
0.009	0.001	0.15	2.25	0.061	2.044
0.009	0.006	0.15	2.25	0.066	1.742
0.006	0.009	0.15	2.25	0.049	1.441

Final selection of initial guesses for BTS retrievals:

R_{Vc}=1.75 µm always

σ_{Vf}=0.4 µm always

σ_{Vc}=0.7 µm always

For AE<0.6

C_{Vf}=0.001 $\mu m^3/\mu m^2$

C_{Vc}=0.009 $\mu m^3/\mu m^2$

R_{Vf1}=0.15 µm

R_{Vf2}=0.225 µm

For AE>1.9

C_{Vf}=0.009 $\mu m^3/\mu m^2$

C_{Vc}=0.001 $\mu m^3/\mu m^2$

R_{Vf1}=0.1 µm

R_{Vf2}=0.15 µm

For AE>=0.6 and AE<1.3 and AOD<0.05 at 500 nm

C_{Vf1}=0.005 $\mu m^3/\mu m^2$

C_{Vc1}=0.005 $\mu m^3/\mu m^2$

R_{Vf1}=0.15 µm

C_{Vf2}=0.006 $\mu m^3/\mu m^2$

C_{Vc2}=0.006 $\mu m^3/\mu m^2$

R_{Vf2}=0.225 µm

For AE>=1.3 and AE<1.9 and AOD<0.05 at 500 nm

C_{Vf}=0.005 $\mu m^3/\mu m^2$

C_{Vc}=0.005 $\mu m^3/\mu m^2$

R_{Vf1}=0.1 µm

R_{Vf2}=0.15 µm

For AE>=0.6 and AE<1.3 and AOD>=0.05 at 500 nm

C_{Vf}=0.009 $\mu m^3/\mu m^2$

C_{Vc}=0.009 $\mu m^3/\mu m^2$

R_{Vf1}=0.15 μm

R_{Vf2}=0.225 μm

For AE>=1.3 and AE<1.9 and AOD>=0.05 at 500 nm

C_{Vf}=0.009 μm^3/ μm^2

C_{Vc}=0.009 μm^3/ μm^2

R_{Vf1}=0.1 μm

R_{Vf2}=0.15 μm

S5. Wildfire episode

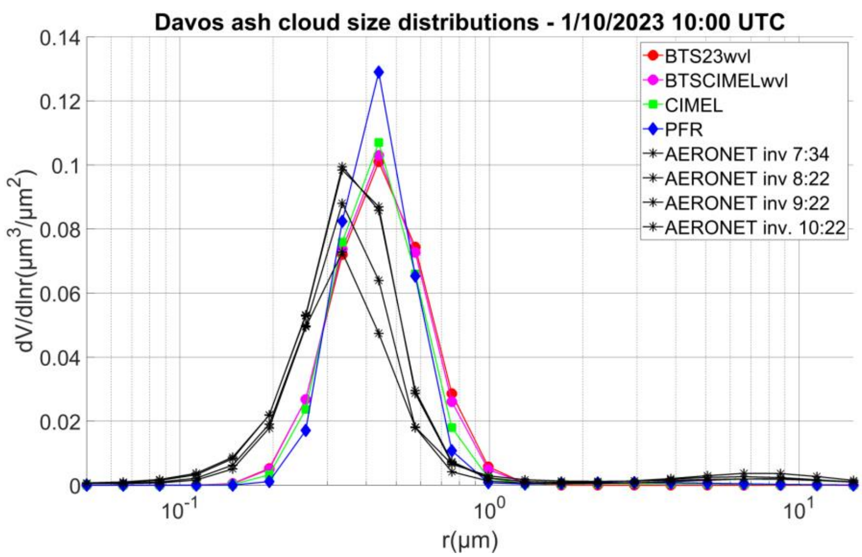

Figure S4: Size distributions from GRASP-PFR, GRASP-BTS and GRASP-CIMEL (coloured lines) for a coincident measurement of AOD and the four closest AERONET size distributions (black lines).

Initial guesses and refractive index:

C_{Vf}:0.007 μm^3/ μm^2

C_{Vc}:0.001 μm^3/ μm^2

R_{Vf}: 0.145 μm

R_{Vc}: 3.154 μm

σ_{Vf}:0.4 μm

σ_{Vc}:0.7 μm

RRI: 1.401

IRI: 0.003

BTS AOD wavelengths:

304, 310, 320, 330, 340, 350, 370, 390, 410, 440, 480, 500, 540, 610, 675, 750, 777, 845, 880, 1020, 1240, 1550 and 1640 nm