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Supplement of

Multidecadal trend analysis of in situ aerosol radiative properties around the world

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Supplementary material:

1) List of instruments characteristic

Table S1 Measurement description

Scattering Instrument, Model number	Scattering wavelengths (nm)[1]	Corrections	Manufacturer Info
TSI total/backscatter nephelometer, 3563	450, 550, 700	Anderson and Ogren [1998]	TSI, Inc. St. Paul, MN USA
Optec open-air nephelometer, NGN-2	550	none applied	Optec, Inc. Lowell, MI USA
Radiance Research nephelometer, M903	532	none applied could use A&O corrections for assumed SAE (Table 4b?) but probably doesn't matter for trends since constant	Radiance Research Seattle, WA, USA
Ecotech Aurora 3000 nephelometer, ...	450, 525, 630	Anderson and Ogren [1998] should use A&O based on Müller et al., 2011	Ecotech Melbourne, Australia
Absorption Instrument, Model number	**Absorption wavelengths (nm)[1]**	**Corrections**	**Manufacturer Info**
Aethalometer AE9, AE10, AE16: white light Aethalometer AE22: 2 wavelengths Aethalometer AE31 and AE33: 7 wavelengths	broadband centered at 840 nm 370 and 880 nm 370,470,520,590,660 ,880,950 nm	C_{ref}=1.80 AE31: C_{ref}=3.5 AE33: C_{ref}=3.0	Magee Scientific, Berkeley, CA USA
PSAP - 1 wavelength PSAP - 3 wavelengths	565^{psap} 467, 530, 660^{psap}	Bond et al. [1999] Ogren [2010]	Radiance Research, Seattle, WA USA
MAAP	637	none applied	Thermo Scientific Franklin, MA USA
CLAP 3w	467, 528, 652	Bond et al. [1999] Ogren [2010]	NOAA Boulder, CO USA

[1]Reported by manufacturer; [psap]In this work we use PSAP data adjusted to 550 nm, based on Bond et al. [1999] for 1- wavelength instrument or Ogren, [2010] for 3-wavelength.

2) Present-day MK results not shown in the paper

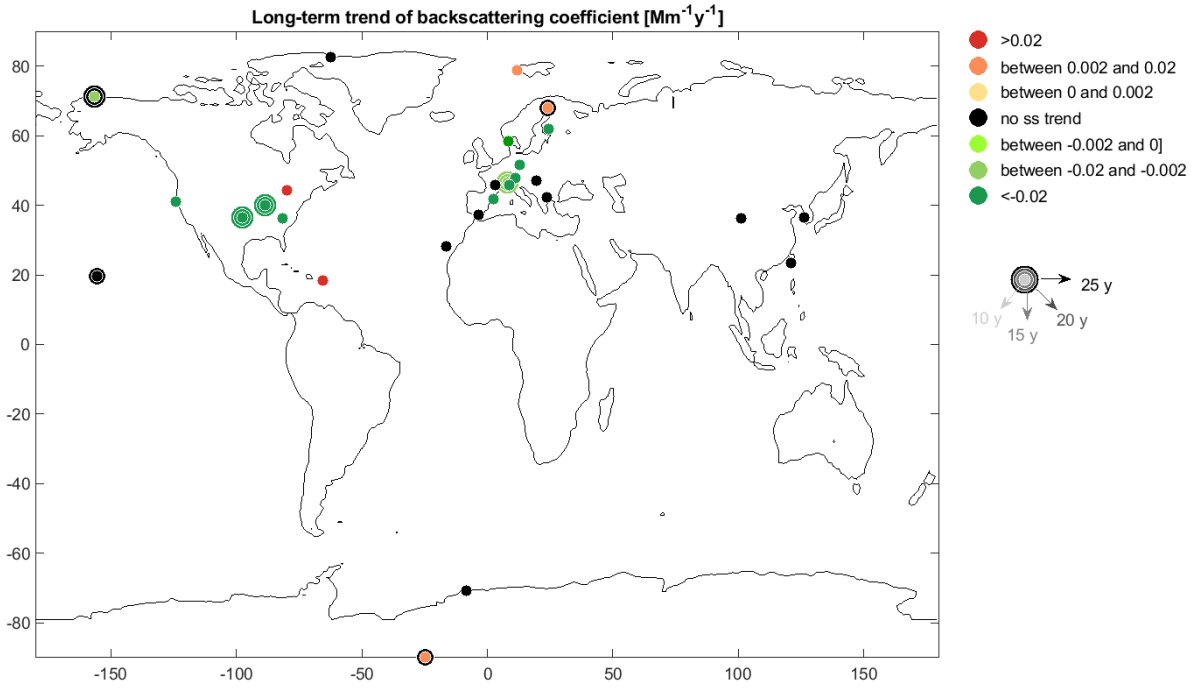

Fig. S1: MK trends results for the backscattering coefficient. Black symbols correspond to stations with no significant trends. Green and orange symbols correspond to statistically significant negative and positive trends, respectively, the magnitude of the trends (slope) being given by the colors as stipulated in the legend. The size of the circles are proportional to the length of the data sets with the central dots representing the present-day 10 years trend ending in 2016, 2017 or 2018. If possible, trends for longer periods were calculated and the larger circles denote the trends for 15 y to 40 y in 5-year increments.

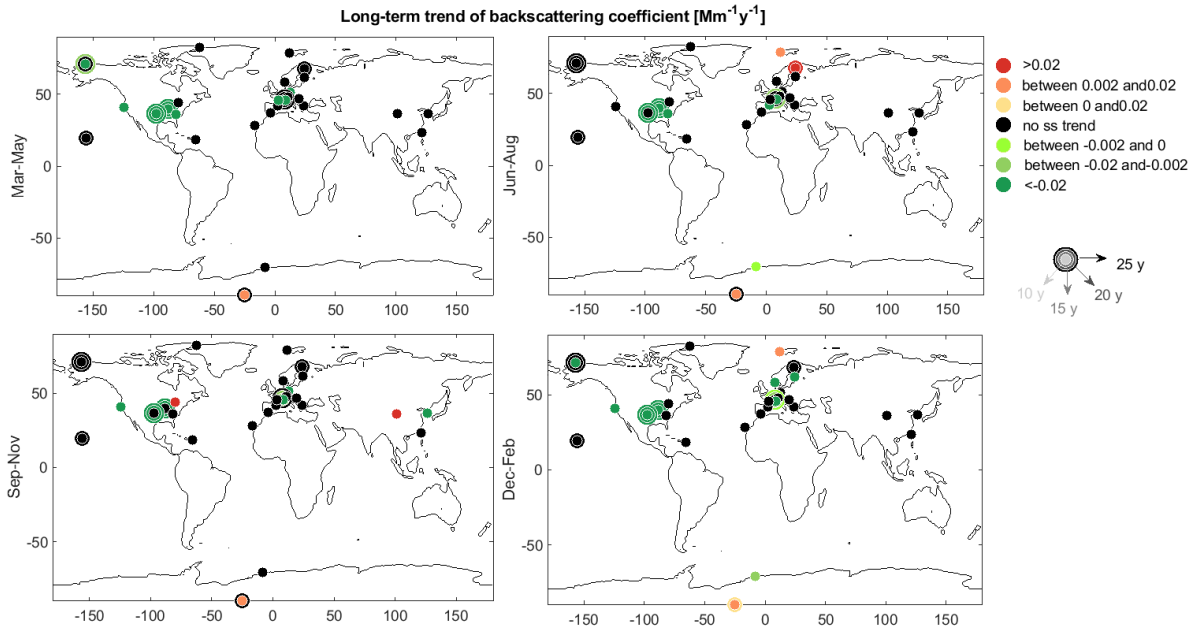

Fig. S2: Seasonal results of the MK trend of the backscattering coefficient. Other details same as Fig. S1.

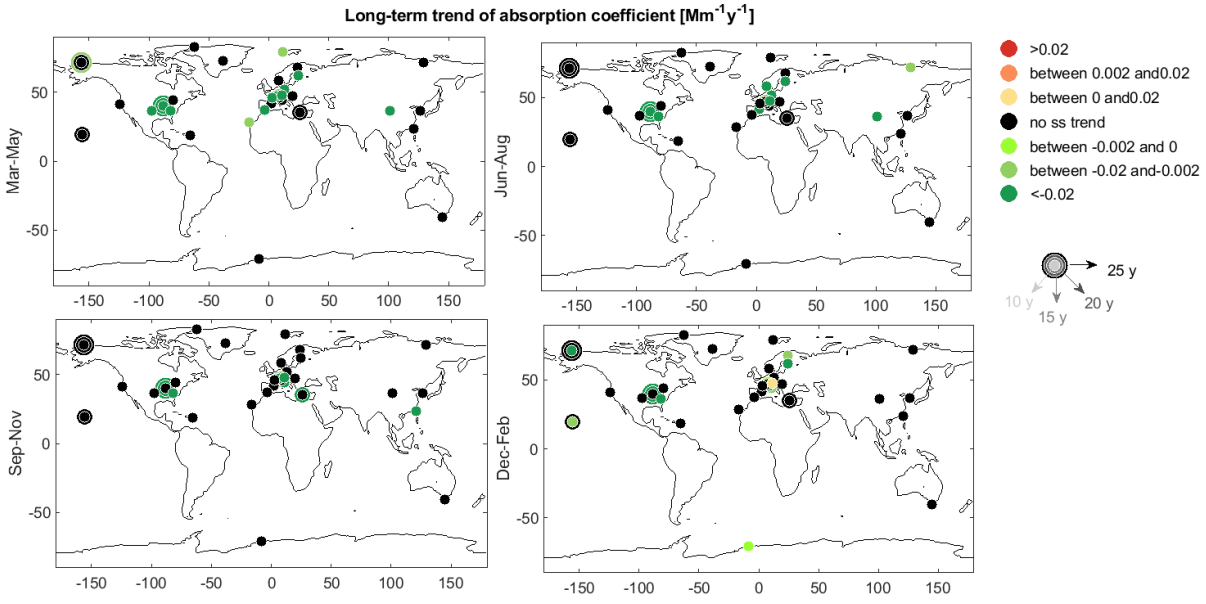

Fig. S3: Seasonal results of the MK trend of the absorption coefficient. Other details same as Fig. S1.

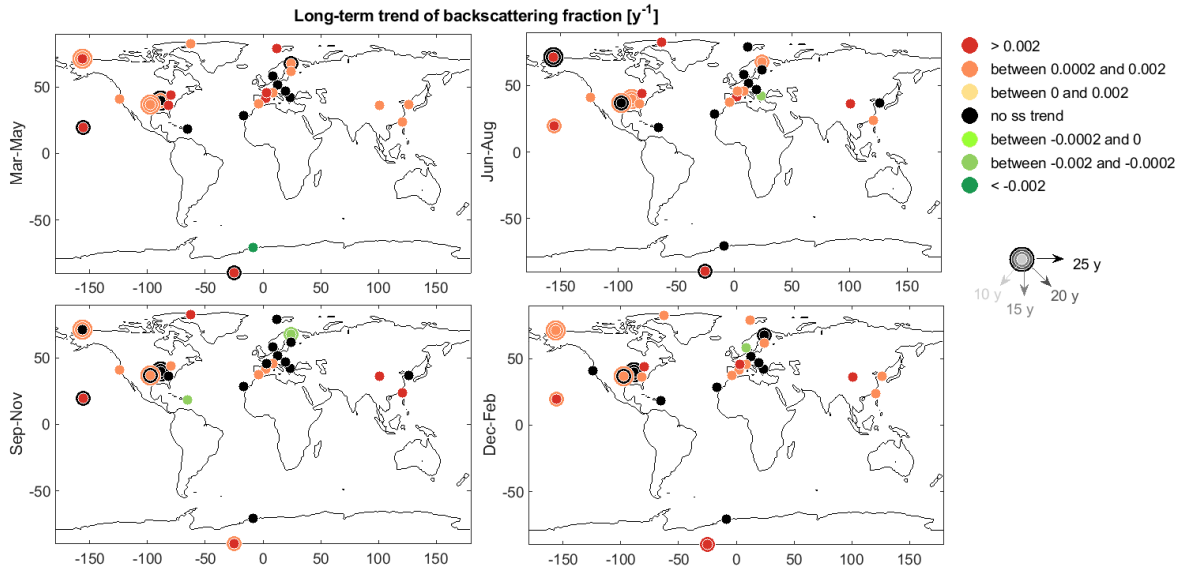

Fig. S4: Seasonal results of the MK trend of the backscattering fraction. Other details same as Fig. S1.

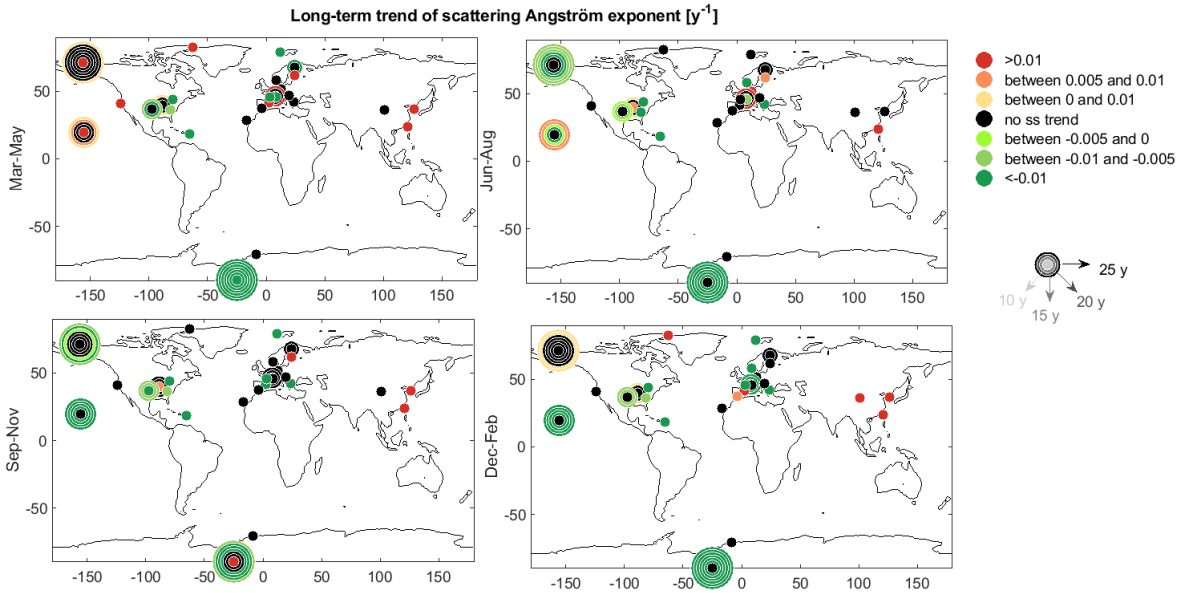

Fig. S5: Seasonal results of the MK trend of the scattering Ångström exponent. Other details same as Fig. S1.

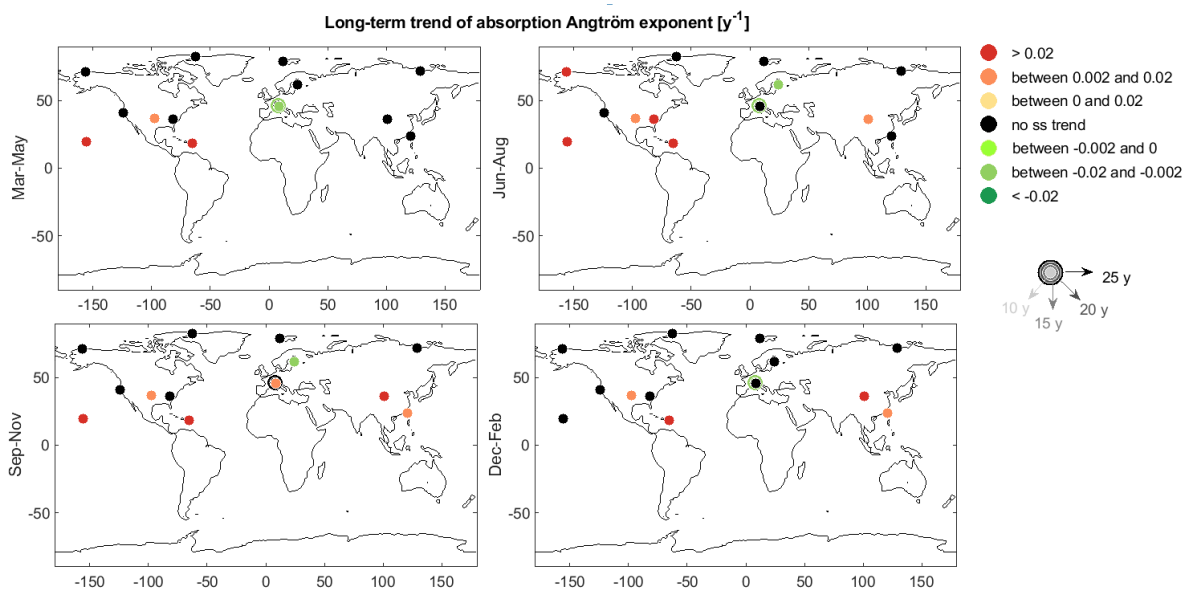

Fig. S6: Seasonal results of the MK trend of the absorption Angström exponent. Other details same as Fig. S1.

3) Present-day trend results for MK, LMS and GLS/ARB methods in %/y

Table S1: Present –day results for MK in %/y

Station	σ_{sp}			σ_{bsp}			σ_{ap}			ω_o			b			$å_{sp}$			$å_{ap}$	
	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y
Africa																				
IZO	-4.60			-2.31			**-3.84**			-0.02			**0.41**			-0.16				
Asia																				
AMY	-1.62			-1.29			-0.27			**-0.08**			0.48			**1.19**				
LLN	-1.94			-1.30			**-7.49**			**0.40**			**1.34**			**1.36**			0.44	
WLG	-1.81			0.51			**-3.22**			**0.05**			**1.86**			1.55			**2.30**	
Europe																				
BEO	-1.26			3.76									**-1.28**			**-1.39**				
BIR	**-3.11**			**-3.27**			**-4.21**			0.11			-0.32			**-1.36**				
CMN							**-1.98**													
FKL							-0.05	**-0.16**												
HPB	**-4.14**			**-3.42**			**-4.41**			**0.06**										
HYY	**-2.91**			**-2.24**			**-4.20**			**0.28**			**0.60**			**0.91**			**-0.44**	
IPR	**-7.30**			**-6.71**			**-1.85**			**-1.04**			**0.86**			**-0.50**			0.11	
JFJ	**-8.34**	**-5.35**	**-2.89**	**-4.71**	**-3.55**	**-2.28**	**-10.22**	**-3.37**		**-0.31**	**-0.11**					**-1.52**	**-0.43**	**0.21**	**-0.73**	**-0.57**
KPS	-1.02			-0.62			-0.50			**0.14**			-0.04			0.00				
MPZ	**-5.47**			**-4.65**			**-5.44**			0.02			-0.06			0.37				
MSY	**-6.00**			**-4.02**			**-1.67**			**-0.41**			**2.16**			**0.51**				
PAL	**2.76**	0.56		**4.09**	0.97		-3.86			**0.26**			0.50	0.01		0.64	0.03			
PAY							**-3.86**													
PUY	-3.24			-1.72			**-3.62**			**0.23**			**1.66**			**-0.97**				
UGR	1.31			1.66			**-0.44**			**0.28**			**0.88**			**0.74**				
ZSF							**-11.79**													
North America																				
ACA	**-4.80**	**-2.64**	**-2.28**																	
APP	**-4.29**			**-3.28**			**-5.35**			**0.08**			**1.34**			**-0.49**			-0.05	
BND	**-3.86**	**-2.45**	**-1.84**	**-3.61**	**-2.30**	**-1.74**	**-2.88**	**-3.17**	**-1.28**	**-0.09**	-0.05	**-0.06**	0.15	**0.43**	**0.18**	**0.44**	0.10	**0.25**		

CPR	**2.93**			**2.59**			**-3.08**			**0.14**			**-0.35**			**-6.34**			**6.73**
EGB	0.93			**2.83**			0.81			0.11			**2.41**			**-1.71**	**-1.40**		
GBN	**-5.42**																		
GLR	1.48																		
HGC	**-3.72**	**-3.39**	**-1.33**																
MCN	**-4.96**	**-3.65**	**-2.40**																
MRN	-0.24	**-0.97**	**-1.74**																
RMN	**-3.57**																		
SGP	**-1.71**	**-1.64**	**-1.67**	**-1.48**	**-1.75**	**-1.31**	-0.59			**-0.19**			**0.92**	**0.27**	**0.50**	**-0.70**	**-0.64**	**-0.53**	**1.79**
SHN	**-2.93**	**-2.76**	**-2.18**																
THD	**-5.16**			**-5.02**			-2.27			**-0.06**			**0.42**			**2.60**			0.35
South Pacific																			
CGO	**3.57**						0.77			0.01									
MLO	**-2.11**	0.01	**0.52**	-0.57	1.02		**-4.59**	1.63		**0.27**	**-0.22**		**3.40**	**0.88**		0.36	**-1.58**	**-0.72**	**6.48**
Polar regions																			
ALT	**-0.23**			-0.04			**2.77**			**0.12**			**1.46**			**1.24**			**1.09**
BRW	**-6.16**	-2.35	**-1.26**	**-3.99**	-1.07	-0.76	**-9.91**	-2.60	-3.53	0.02	-0.05	-0.03	**1.72**	**1.08**	**0.54**	**1.69**	0.68	**-0.27**	-0.54
NMY	0.97			-0.51			**-1.76**			0.01			-0.62			-0.61			
SPO	**2.59**	-0.22	**1.80**	**8.08**	0.53								**6.10**	0.34		-1.66	**-1.91**	**-2.41**	
SUM							-0.36	1.58											
TIK							**-0.90**												-0.28
ZEP	**4.32**			**4.00**			-0.19			**0.12**			**0.41**	**0.41**		**-1.78**	**-0.29**	**-1.78**	**0.38**

Table S2: Present –day results for GLS/ARB applied on daily median in %/y

Station	σ_{sp}			σ_{bsp}			σ_{ap}			ω_o			b			$å_{sp}$			$å_{ap}$	
	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y
Africa																				
IZO	11.30			11.76			0.48			0.10			0.45			-2.04				
Asia																				
AMY	**-3.11**			**-2.59**			**-1.52**			**-0.06**			0.27			**1.01**				

LLN	-5.39			-2.39			**-10.48**			**0.57**			**2.52**			**1.53**			1.12
WLG	**-7.45**			-2.10			**-6.57**			**0.08**			**3.38**			0.12			**3.30**
Europe																			
BEO	**-4.69**			0.12									**-1.47**			**-2.16**			
BIR	**-6.49**			**-6.09**			**-16.84**			-0.04			-0.30			**-2.35**			
CMN							**-7.80**												
FKL							0.77	**-1.69**											
HPB	**-6.67**			**-4.46**			**-8.15**			**0.15**									
HYY	**-5.90**			**-2.96**			**-10.35**			**0.48**			**0.70**			**0.97**			**-0.63**
IPR	**-18.89**			**-15.80**			**-9.91**			**-1.53**			-0.13			**-0.54**			0.19
JFJ	**-24.47**	**-12.29**	**-6.60**	**-12.56**	**-7.61**	**-5.37**	**-14.23**	**-7.76**		**-0.36**	**-0.15**					**-2.01**	**-0.88**	-0.11	**-1.20**	**-0.53**
KPS	-0.42			-0.67			-0.90			**0.20**			-0.03			-0.20			
MPZ	**-7.60**			**-5.68**			**-9.25**			**0.12**			-0.38			0.15			
MSY	**-12.14**			**-6.18**			**-3.18**			**-0.44**			**4.60**			**2.08**			
PAL	**10.59**	1.44		**11.67**	**2.72**		-5.55			**0.55**			**0.96**	**0.83**		0.36	-0.23		
PAY							**-4.04**												
PUY	**-5.39**			-2.15			**-5.18**			**0.50**			**3.14**			**-2.03**			
UGR	-0.92			0.49			**-2.44**			**0.41**			**1.22**			**0.88**			
ZSF							**-20.45**												
North America																			
ACA	**-9.41**	**-7.62**	**-5.49**																
APP	**-5.92**			**-3.51**			**-6.35**			**0.11**			**1.50**			**-0.68**			**1.52**
BND	**-6.54**	**-4.67**	**-3.06**	**-5.36**	**-3.41**	**-2.48**	**-3.70**	**-4.57**	**-2.04**	**-0.13**	-0.02	-0.01	**0.38**	**0.54**	**0.15**	**0.43**	0.03	**0.22**	
CPR	**2.57**			**2.35**			-5.37			**0.23**			**-0.27**			**-9.80**			**8.05**
EGB	-2.61			1.87			**3.28**			-0.03			**2.58**			**-2.34**	**-2.22**		
GBN	**-5.84**																		
GLR	1.99																		
HGC	**-7.48**	**-5.49**	**-2.91**																
MCN	**-7.06**	**-7.11**	**-5.09**																
MRN	-0.55	**-1.04**	**-1.95**																
RMN	-1.44																		
SGP	**-3.40**	**-3.61**	**-3.09**	**-2.87**	**-3.13**	**-2.06**	**-1.38**			**-0.12**			**0.88**	**0.30**	**0.57**	**-1.03**	**-0.93**	**-0.72**	**2.44**
SHN	**-6.82**	**-8.41**	**-6.11**																
THD	**-6.78**			**-5.96**			**-10.62**			-0.06			**0.94**			**3.01**			0.50

South Pacific																						
CGO	**3.98**						6.49			**0.06**												
MLO	**-15.20**	-0.77	**2.59**	**-5.64**	0.34		**-18.98**	2.84		**0.31**	**-0.37**		**3.96**	**0.86**		0.03	**-2.15**	**-1.84**	**9.23**			
Polar regions																						
ALT	**-5.36**			**-3.39**			**-9.06**			**0.23**			**3.31**			**1.08**			1.07			
BRW	**-7.13**	-1.44	-0.65	**-3.60**	-0.08	0.12	**-11.33**	-0.94	**-3.00**	**0.14**	0.12	0.03	**5.00**	-2.31	0.05	**1.82**	**-1.03**	**-1.40**	1.15			
NMY	42.10			295.99			**-5.07**			**0.02**			**-1.38**			**-1.10**						
SPO	0.52	-2.44	1.24	**8.27**	0.06								**7.84**	**1.12**		**-1.77**	**-1.62**	**-1.96**				
SUM							-5.20	0.22														
TIK							0.25												-0.38			
ZEP	**6.14**			**7.68**			0.78			0.13			**1.67**	**1.67**		**-1.94**	-0.86	**-1.94**	0.62			

Table S3: Present –day results for LMS applied on the logarithm of monthly median in %/period

Station	σ_{sp}			σ_{bsp}			σ_{ap}			ω_o			b			$å_{sp}$			$å_{ap}$	
	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y	20 y	10 y	15 y
Africa																				
IZO	-0.92			0.71			-2.95			-0.10			0.07			-10.75				
Asia																				
AMY	-0.47			**-0.89**			-1.16			-1.46			0.08			**2.32**				
LLN	-1.10			-2.71			**-76.35**			**5.72**			**0.82**			**3.04**			11.88	
WLG	**-0.88**			0.12			**-3.77**			1.16			**1.78**			6.64			**6.54**	
Europe																				
BEO	**-1.91**			-4.70									-0.60			**-4.52**				
BIR	-1.75			**-140.52**			**-65.36**			0.06			**-0.31**			**-10.06**				
CMN							-20.90													
FKL							0.60	**-3.17**												
HPB	**-1.75**			**-4.70**			**-6.87**			0.52										
HYY	**-1.76**			**-7.53**			**-24.11**			**3.71**			**0.47**			**1.46**			**-20.84**	
IPR	**-3.31**			**-6.65**			**-2.68**			**-6.45**			**0.53**			-0.45			4.25	
JFJ	**-12.60**	**-6.43**	**-2.97**	**-6.86**	**-4.31**	**-2.52**	**-3.71**	**-2.88**		**-3.60**	**-1.62**					**-3.34**	-1.16	-0.03	-23.61	**-0.57**
KPS	0.02			-0.09			-0.48			1.34			-0.10			-0.49				
MPZ	**-1.33**			**-2.60**			**-4.56**			**1.48**			-0.21			0.11				

MSY	**-2.18**			**-3.02**			-2.05			**-5.73**			**1.80**			9.04			
PAL	**2.51**	0.79		**8.64**	2.12		-1.04			**5.55**			0.00	-0.04		-2.82	-1.20		
PAY							**-1.54**												
PUY	-0.83			-1.92			**-1813.71**			2.15			**0.59**			**-2.69**			
UGR	-0.17			0.11			-0.68			1.24			**0.62**			1.08			
ZSF							**-23.63**												
North America																			
ACA	**-2.21**	**-1.45**	**-1.01**																
APP	**-1.58**			**-2.89**			**-7.44**			**1.13**			**0.80**			**-0.82**			**14.80**
BND	**-1.47**	**-1.04**	**-0.72**	**-3.35**	**-2.13**	**-1.49**	**-3.63**	**-3.42**	**-1.37**	**-1.93**	-0.30	-0.39	0.22	**0.30**	0.10	**0.71**	0.13	**0.46**	
CPR	**0.78**			**2.42**			-11.68			**6.25**			-0.06			**-10.55**			19.42
EGB	-0.06			**3.16**			**15.18**			-0.52			**1.56**			**-3.25**	**-3.00**		
GBN	**-4.21**																		
GLR	0.58																		
HGC	**-3.64**	**-2.76**	**-1.36**																
MCN	**-1.54**	**-1.36**	**-1.04**																
MRN	-0.03	-0.32	**-0.59**																
RMN	**-3.04**																		
SGP	**-1.04**	**-0.86**	**-0.58**	**-2.30**	**-2.10**	**-1.10**	-2.06			-1.17			**0.56**	**0.17**	**0.32**	**-1.32**	**-1.35**	**-1.06**	**7.01**
SHN	**-1.88**	**-1.68**	**-1.16**																
THD	**-1.99**			**-6.43**			-4.95			-1.44			**0.36**			**19.33**			3.87
South Pacific																			
CGO	**2.21**						-0.59			**5.51**									
MLO	**-32.28**	2.54	6.45	-0.08	**0.90**		**-2.81**	**1.72**		3.59	**-3.58**		**1.75**	**0.60**		-3.00	**-8.07**	**-4.40**	**26.87**
Polar regions																			
ALT	**-2.57**			-3.13			-1.93			1.25			**0.86**			3.42			9.69
BRW	**-3.32**	0.01	0.21	**-10.06**	1.93	2.75	**-2.76**	1.63	0.30	2.94	13.80	2.20	**1.32**	**0.47**	**0.20**	1.11	-9.69	**-22.49**	5.28
NMY	3.73			0.18			**-0.81**			2.94			**-0.86**			-45.17			
SPO	1.55	-0.27	**2.87**	**3.17**	0.22								**3.69**	0.29		**-5.89**	**-5.46**	**-4.99**	
SUM							-0.61	-0.58											
TIK							-14.66												-4.37
ZEP	**5.72**			**4.75**			0.39			**3.49**			0.22	0.22		-3.65	-1.11	-3.65	0.78

4) Evolution of 10y trends

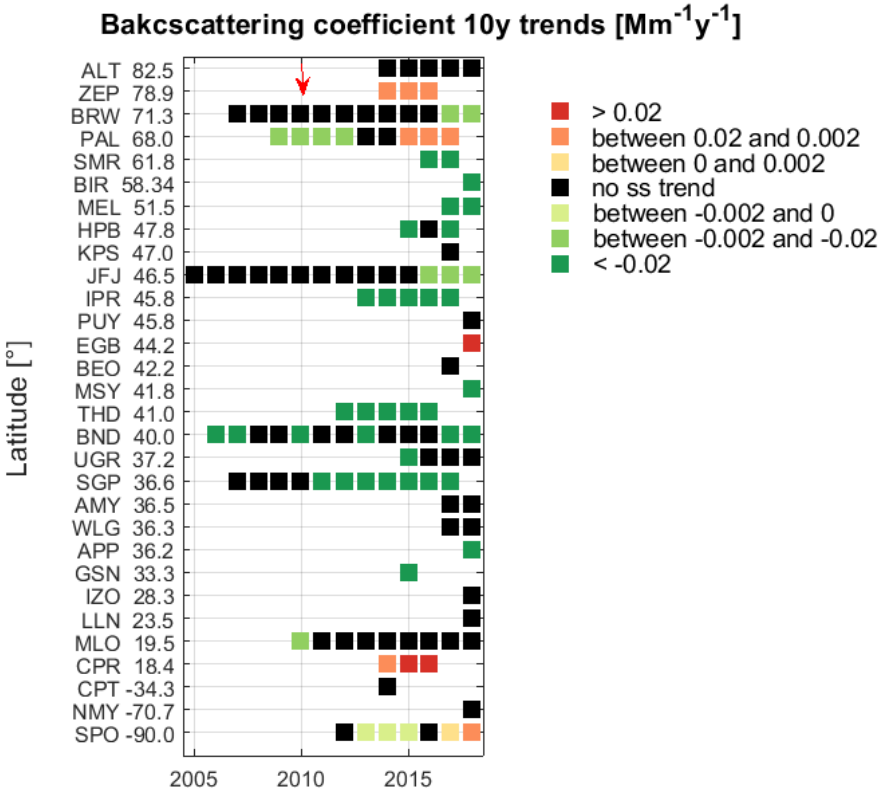

Fig. S7: Time series of 10y backscattering coefficient trends as a function of station latitude.

5) RH trends

Table S4: MK present-day trends in %/y of RH measured by the nephelometer. The ss trends are given in bold.

	MK 10y	MK 20y	MK 30y
ACA	0.00	0.04	0.04
APP	**-0.34**	**-0.33**	
BND	-0.08	0.11	**0.11**
CPR	**0.08**		
EGB	**-0.51**		
GBN	-0.25		
GLR	-0.03		
MCN	-0.06	0.02	0.06
MRN	**0.08**	0.00	-0.03
RMN	0.04		
SHN	**0.31**	**0.15**	**0.13**

THD	**-0.30**		
HGC	-0.18	-0.12	-0.02
BEO	-0.23		
BIR	**-0.13**		
HPB	0.03		
HYY	**0.12**		
IPR	-0.17		
MSY	-0.18		
PAL	0.14	0.09	
PUY	**0.30**		
UGR	**-0.55**		
AMY	**0.56**		
LLN	**-1.80**	**-1.88**	
ALT	0.00		
BRW	0.04	**0.05**	
IZO	**-0.72**		
CGO	-0.02		

6) Relation between SSA trends and absorption and scattering trends

The SSA trends depend directly on σ_{sp} and σ_{ap} trends. A σ_{ap} trend expressed in %/y larger (lower) than the σ_{sp} trend will result in a decreasing (increasing) SSA trend, respectively (see Fig. S7). In contrast, when the σ_{ap} trend (in %) is lower than the σ_{sp} trend an increasing SSA trend will result. The cyan and green curves in Fig. 7 give an idea of the variability of the SSA trend corresponding to Aethalometer measurement error (30%) as well as to the potential C_{ref} errors for continental background aerosol (about 30% for C_{ref} between 2.5 and 4.5). The figure suggests that higher σ_{ap}/σ_{sp} ratios lead to larger SSA trends for similar σ_{ap} and σ_{sp} trends. However, the simple rule still holds that when the σ_{ap} trend is larger than the σ_{sp} trend, the SSA trend will be decreasing (i.e., the aerosol will be getting darker).

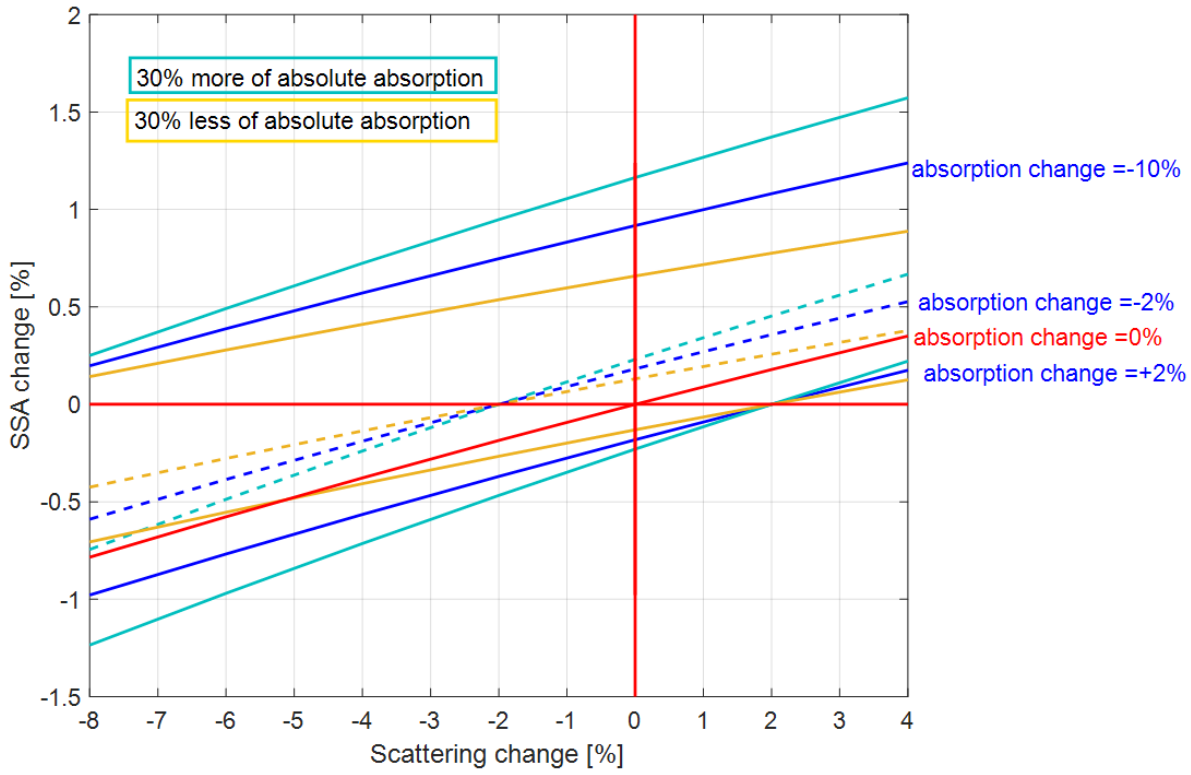

Figure S8: Single scattering albedo change as a function of the scattering coefficient change for several absorption coefficient changes. The change are always given in %. The σ_{ap}/σ_{sp} for the dark blue lines is of 0.1. The green and cyan lines represent the same curves but for an increase of absorption of +30% and -30% respectively, corresponding to σ_{ap}/σ_{sp} equal to 0.133 and 0.067. The chosen σ_{ap} and σ_{sp} trend ranges correspond to the results of this study and the SSA trend range of this study is of -1% to 0.4%.