Papers

Seasonal changes of the aerosol optical thickness for the atmosphere over the Baltic Sea – preliminary results*

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Aerosol optical thickness
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Abstract

A statistical analysis of the aerosol optical thickness and the Ångström parameters derived from measurements of spectral solar radiation at the surface of the southern and western Baltic Sea is presented. The experimental data were collected on cloudless days during over 20 cruises from March to September within a 4-year period (1994–1998). The monthly and seasonal variability of the aerosol optical thickness for two wind direction sectors – northern sector 270°–N–90° and southern sector 90°–S–270° – is analysed.

1. Introduction

Meteorological conditions over the Baltic Sea are highly variable. Various air masses – arctic, polar and tropical in their maritime and continental variants – may advect into the Baltic Sea area. Moreover, the atmosphere over this region is influenced by the surrounding land. One effect of the land-locked location of the Baltic Sea is that the atmosphere over the sea is neither typically maritime nor continental. This very complex situation also affects aerosols.

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The quantity which characterises the cumulative attenuation of solar radiation by aerosols at a given wavelength in the whole column of the atmosphere over a given area is the aerosol optical thickness. This depends directly on the particle concentrations, size distributions, compositions and vertical profiles of aerosols, and indirectly on their sources and history. On the way from their sources aerosols can change owing to transport processes (turbulent diffusion, advection, vertical convection in the atmosphere), dry and wet deposition, chemical and physical transformation, and coagulation. Moreover, the sizes of hygroscopic components such as sea salt particles depend on the ambient relative humidity (Gong *et al.* 1987).

The influence of various atmospheric conditions on the values of the aerosol optical thickness have been studied previously by many researchers (*e.g.* Peterson *et al.* 1981, Smirnov *et al.* 1994, Villevalde *et al.* 1994, Russak 1996). In particular, the aerosol optical thickness for the atmosphere over the Baltic Sea has been discussed by *e.g* Eerme (1983), Weller & Leiterer (1988), Villevalde *et al.* (1989), Gulyaev *et al.* (1990), Smirnov *et al.* (1995), Kuśmierczyk-Michulec & Darecki (1996), Kuśmierczyk-Michulec *et al.* (in press). The influence of aerosols on solar radiation transmittance in the Baltic Sea region has been investigated by Krężel (1985, 1992). Some measurements on the Baltic Sea coast were considered by Shifrin *et al.* (1980), von Hoyningen-Huene & Wendisch (1994), and Persson (1999). Table 1 summarises the known measurements of the aerosol optical thickness carried out on the Baltic Sea or on its coast. The results presented by Weller & Leiterer (1988), Gulyaev *et al.* (1990), von Hoyningen-Huene & Wendisch (1994) and Smirnov *et al.* (1995) occur in association with a traditional air mass classification that assumes the existence of two main fronts (polar and arctic), and thus, three air mass types (arctic, polar and tropical) with their maritime and continental variants. The classification of aerosol types presented by Kuśmierczyk-Michulec *et al.* (in press) is based on the travelling time of air masses over the sea surface.

However, these studies are based on a very limited number of measurements or on data from coastal stations. In this work we investigate the aerosol optical thickness spectra obtained for measurements taken in a cloudless maritime atmosphere over the southern and northern Baltic Sea, from March to September over a 4-year period. Such an empirical data set creates an opportunity to analyse the seasonal variability of the Baltic aerosol optical properties expressed in terms of the aerosol optical thickness.

Since for the majority of our data detailed information on the air mass types and the air mass trajectories was not available, the wind direction was used as a rough indicator of air mass type. Our data were separated into

Table 1. Summary of the aerosol optical thickness over the Baltic Sea

References	Air mass type	Area	N	N_{days}	$\tau_A(550)$	σ	α	σ_α
Shifrin *et al.*(1980)	–	Baltic Sea coast	–	2	0.20	–	–	–
Eerme (1983)	–	Baltic Sea	–	9	0.17	–	–	–
Weller & Leiterer (1988)	continental	Baltic Sea	1	1	0.62	–	–	–
	maritime	Baltic Sea	1	1	0.18	–	–	–
	–	Baltic Sea	17	–	–	–	1.17	0.26
	–	Baltic Sea coast, Germany	17	–	–	–	1.25	0.16
Villevalde *et al.* (1989)	–	Baltic Sea	–	5	0.23	–	–	–
Gulyaev *et al.* (1990)	continental polar	Baltic Sea	1	1	0.18	–	0.90	–
Von Hoyningen–Huene & Wendisch (1994)	continental polar	Baltic Sea coast Zingst, Germany	–	–	0.21	–	1.17	–
	continental polar (aged)		–	–	0.20	–	1.06	–
	continental tropical		–	–	0.29	–	1.12	–
	continental tropical (aged)		–	–	0.24	–	0.92	–
	maritime tropical		–	–	0.10	–	0.42	–
	maritime polar		–	–	0.11	–	0.46	–
Smirnov *et al.* (1995)	continental polar	Baltic Sea (May, July 1984)	1	1	0.46	–	1.14	–
	maritime arctic		1	1	0.09	–	0.99	–
	maritime polar (modified)		1	1	0.45	–	1.37	–
	centre of a high pressure system		3	–	0.12	0.04	0.77	0.10

Table 1. (continued)

References	Air mass type	Area	N	N_{days}	$\tau_A(550)$	σ	α	σ_α
Kuśmierczyk-Michulec & Darecki (1996)	–	Baltic Sea (April, May, June, August 1994)	66	19	0.25	0.14	–[a]	–[a]
Kuśmierczyk-Michulec *et al.* (in press)	continental	Baltic Sea (July 1997)	58	13	0.19	0.07	1.20	0.22
	maritime		22	4	0.35	0.17	0.39	0.11
	continental-maritime		65	10	0.17	0.09	1.27	0.38
Persson (1999)	–	Baltic Sea coast Sweden (1983–1997)						
		Stockholm	–	–	0.16	0.10	1.3[b]	–
		Norrköping	–	–	0.14	0.10	1.3[b]	–
		Visby (Gotland)	–	–	0.16	0.11	1.3[b]	–
		Lund	–	–	0.18	0.13	1.3[b]	–

N – number of spectra analysed,
N_{days} – number of measurement days,
σ – standard deviation of the aerosol optical thickness,
α – Ångström parameter,
σ_α – standard deviation of the Ångström parameter,
(–) – indicates that no information is available,
[a]/indicates that spectra of the aerosol optical thickness are not well approximated by the Ångström power law,
[b]/indicates that the wavelength exponent α is set constant.

two classes, corresponding to two wind direction sectors: a northern sector 270°–N–90° (arctic and polar air masses) and a southern sector 90°–S–270° (polar and tropical air masses).

2. Measurements

Two kinds of irradiance data were used in this study, depending on the method applied to derive the aerosol optical thickness. In the standard method, the total and diffuse solar spectral irradiances were measured simultaneously by means of a shadow-band spectrophotometer for eight spectral channels (each channel of 10 nm width): 412 nm, 490 nm, 510 nm, 555 nm, 670 nm, 765 nm and 865 nm. The precision of the measurements was estimated at 2–3% for the total and 5–7% for the diffuse irradiances. The method of measurements has been described in recent publications (Olszewski *et al.* 1995, Kuśmierczyk-Michulec & Darecki 1996). Irradiances averaged over 1 minute were used in calculations.

In the pyranometric method, the broad-band downward irradiance was measured by means of two Eppley Precision Spectral Pyranometers fitted with hemispherical Schott filters (GG395 and RG695 – the numbers denote cut-off wavelengths in nm). The measurement error, comprising all the error sources of the actinometric measurements on the ship, did not exceed 3–5%. The 5-minute means of irradiance were used in the calculations.

Table 2. Meteorological conditions during measurements

Month	Meteorological conditions								
	T [°C]		RH [1/100]		p [hPa]		U [m s^{-1}]		Wind
	min	max	min	max	min	max	min	max	direction
March	–1.6	4.7	0.50	0.94	995	1042	3	11	38°–S–290°
April	1.9	5.5	0.67	0.90	1010	1023	4	9.4	10°–S–350°
May	6.9	15	0.61	0.97	997	1027	0.5	13	3°–S–310°
June	8	14	0.83	0.96	1010	1018	3	6.8	180°–S–250°
July	14.6	22	0.52	0.99	1013	1017	1.2	12	1°–S–352°
August	14.2	24	0.68	0.97	1009	1020	0	7.2	90°–S–340°
September	8.7	22	0.54	0.99	1001	1021	1.5	9.5	20°–S–250°
November	–0.6	0.8	0.54	0.76	1026	1027	4.5	5	120°

T – air temperature,
RH – relative humidity,
p – air pressure,
U – wind speed measured at a height of 10 m over the sea surface.

The irradiance measurements were accompanied by the standard meteorological observations. The meteorological conditions during the measurements are summarised in Table 2, while the locations of the measurement stations are shown in Fig. 1.

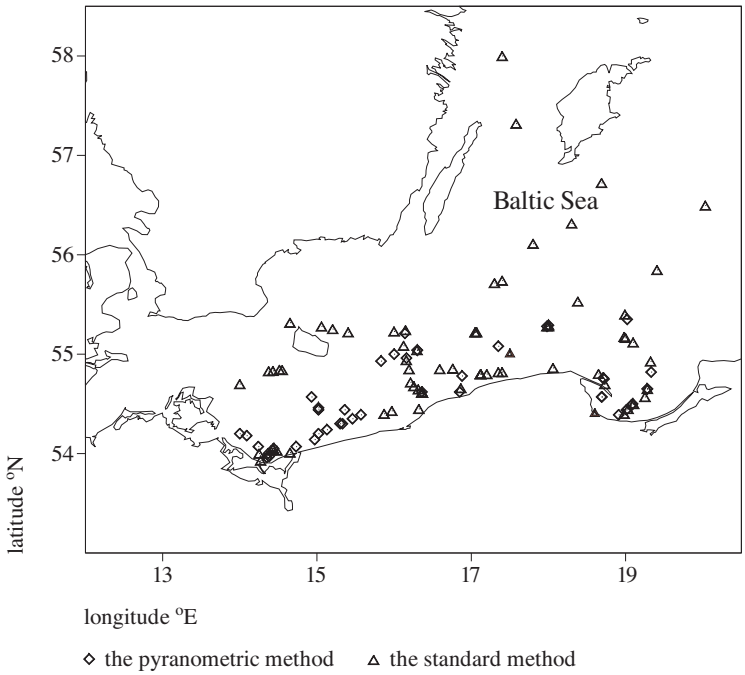


Fig. 1. The locations of the measurement stations

3. Methods

3.1. Standard method

In the standard method the aerosol optical thickness $\tau_A(\lambda)$ was obtained from the following expression:

$$\tau_A(\lambda) = m^{-1} \ln T^{-1}_{A,\,dir}(\lambda), \tag{1}$$

where m is the atmospheric optical mass (Kasten 1966);
the transmittance function for aerosol extinction $T_{A,dir}$ is defined as

$$T_{A,\,dir}(\lambda) = \frac{E^{emp}_{d,\,tot}(\lambda) - E^{emp}_{d,\,dif}(\lambda)}{F_s(\lambda)\, T_R(\lambda)\, T_{O_3}(\lambda)\, T_W(\lambda)\, T_U(\lambda)\, \cos\theta}, \tag{2}$$

where θ is the solar zenith angle, and $E^{emp}_{d,\,tot}(\lambda)$, $E^{emp}_{d,\,dif}(\lambda)$ are the respective total and diffuse measured narrow-band downward irradiances at the sea surface, T_R, T_W, T_{O_3} and T_U are the respective transmittance functions for Rayleigh scattering (van Stokkom & Guzzi 1984), water vapour absorption, ozone absorption and uniformly mixed gas absorption (Leckner 1978, Bird & Riordan 1986). The ozone amount was assumed to be in accordance with the long-term monthly means from the Belsk Geophysical Observatory (Dziewulska-Łosiowa 1991). Values of the mean extraterrestrial spectral irradiance $F_s(\lambda)$ are based on Neckel & Labs (1984) and corrected for the Earth–Sun distance after Paltridge & Platt (1976).

The total statistical (random) error in the zenithal aerosol optical thickness obtained by the standard method was estimated at about $\pm\,0.02$–0.04, the systematic error at about 0.02 (Rozwadowska & Kuśmierczyk-Michulec 1998).

3.2. Pyranometric method

In the pyranometric method (Rozwadowska & Kuśmierczyk-Michulec 1998) the real southern Baltic aerosol is assumed to be a mixture of the model aerosols by McClatchey *et al.* (1984), which have strictly defined optical properties (absorption coefficient c and scattering coefficient b, asymmetry parameter of the scattering function g). The extinction coefficients given in McClatchey *et al.* (1984) are normalised to $1\,\mathrm{km}^{-1}$ at $\lambda = 550\,\mathrm{nm}$. Thus, the total aerosol optical thickness is defined as

$$\tau_A(\lambda) = \sum_{i=1}^{3} \tau_{A,\,i}(550)\,\frac{c_{A,\,i}(\lambda)}{c_{A,\,i}(550)}, \tag{3}$$

where

i – model aerosol type by McClatchey *et al.* (1984): 1 – maritime, 2 – continental, 3 – stratospheric.

The optical thickness of the stratospheric aerosol is assumed to be constant $\tau_{A,\,3}(550) = 0.0047$. The optical thickness of the continental and maritime components at 550 nm can be retrieved by numerical solution of the following set of equations:

$$\begin{aligned} E^{emp}_{d,\,tot}(VIS) &= E^{emp}_{d,\,tot}(\mathrm{GG395}) - E^{emp}_{d,\,tot}(\mathrm{RG695}) = \\ &= \int_{295}^{2800} (f_{\mathrm{GG395}}(\lambda) - f_{\mathrm{RG695}}(\lambda)) \times \\ &\times\; E^{mod}_{d,\,tot}(\lambda,\, \tau_{A,\,1}(550),\, \tau_{A,\,2}(550))\, d\lambda, \end{aligned}$$

$$E^{emp}_{d,\,tot}(IR) = E^{emp}_{d,\,tot}(\mathrm{RG695}) = \\ = \int_{295}^{2800} f_{\mathrm{RG695}}(\lambda)\, E^{mod}_{d,\,tot}(\lambda,\, \tau_{A,\,1}(550),\, \tau_{A,\,2}(550))\, d\lambda, \quad (4)$$

where
$E^{emp}_{d,\,tot}$ – measured broad-band total downward irradiance at the sea surface,
$f_{\mathrm{GG395}}(\lambda)$, $f_{\mathrm{RG695}}(\lambda)$ – Schott filter transmittance factors.

The modelled total downward irradiance $E^{mod}_{d,\,tot}$ is expressed as

$$E^{mod}_{d,\,tot}\ (\lambda, \tau_{A,\,1}(550),\, \tau_{A,\,2}(550)) = \\ = F_s(\lambda)\, T_{\mathrm{atm}}(\lambda,\, \theta,\, p,\, w(e),\, T,\, O_3,\, \tau_{A,\,1}(550),\, \tau_{A,\,2}(550))\, \cos\theta, \quad (5)$$

where
T_{atm} – irradiance transmittance for the homogeneous atmosphere computed using the δ approximation of the scattering function and the single scattering radiative transfer model (quasi-single scattering model) (Woźniak *et al.* 1996),
T – air temperature,
O_3 – amount of atmospheric ozone [atm–cm],
$w(e)$ – precipitable water vapour in a vertical path [cm],
e – surface water vapour pressure [hPa],
p – atmospheric pressure [hPa].

This method is encumbered with a large systematic error which, however, may be corrected by the following relation:

$$\tau^{dir}_A(\lambda) = a_1(\lambda)\, \tau^{pyr}_A(\lambda), \quad (6a)$$

where the coefficient $a_1(\lambda)$ for the wavelength in the 412 nm to 865 nm range can be approximated by the polynominal

$$a_1(\lambda) = 2.339 - 2.713\times10^{-3}\lambda + 1.358\times10^{-6}\lambda^2. \quad (6b)$$

The above equation is based on the comparison between the results obtained pyranometrically and directly (Rozwadowska & Kuśmierczyk-Michulec 1998).

After correction of the systematic error, the statistical (random) error in the estimation of optical thickness by means of the pyranometric method is about ± 0.06.

4. Results

Automatic data acquisition enabled the collection of a large data set, which included continuous measurements of the solar spectral direct component (standard method) or the broad-band total downward irradiance

(pyranometric method). On the basis of these measurements, spectra of the hourly mean values of the aerosol optical thickness were calculated. The application of the linear relationship between the pyranometric and standard methods (eq. (6)) enabled all data to be treated equally.

In order to illustrate the seasonal changes occurring in the atmosphere, the mean values of the aerosol optical thickness $< \tau_A(555) >$ as well as their standard deviations σ_τ for the consecutive months were calculated (Table 3).

Table 3. Monthly variability in the aerosol optical thickness and the Ångström parameters for λ in the 412 nm to 865 nm range

Month	N	N_{days}	$< \tau_A(555) >$	σ_τ	$< \tau_A(\lambda) >$	
					γ	α
March	29	7	0.111	0.067	0.072	0.77
April	31	7	0.090	0.080	0.057	0.89
May	87	19	0.197	0.108	0.115	0.95
June	13	5	0.209	0.093	0.122	0.86
July	117	13	0.215	0.112	0.126	0.92
August	41	11	0.233	0.100	0.130	1.04
September	53	10	0.236	0.114	0.131	1.05
November	5	1	0.110	0.023	0.067	0.88

N – the number of cases,
N_{days} – the number of days,
γ, α – were calculated on the basis of eq. (7) for the spectral values of the aerosol optical thickness averaged over a month.

Additionally, the analysis of the aerosol optical thickness for λ in the 412 nm to 865 nm range in the context of the Ångström parameters γ and α (*e.g.* Bokoye *et al.* 1997):

$$\tau_A(\lambda) = \gamma \left(\frac{\lambda}{1000}\right)^{-\alpha}, \tag{7}$$

is included in Table 3. It should be mentioned that the mean values $< \tau_A(\lambda) >$ are very well represented by the usual Ångström power law (the correlation coefficient 0.99). However, it was found that single measurements might diverge from it.

The lowest values of the mean aerosol optical thickness (0.09–0.11) were recorded in March, April and November. The highest values were obtained in August (0.23 ± 0.10) and September (0.24 ± 0.14). This conclusion may be regarded as a confirmation of the results given by Rozwadowska & Isemer (1998), where the minimum zenithal aerosol transmittances for irradiance

(maximum aerosol optical thickness) calculated for the southern and western Baltic were obtained for July, August and September. Moreover, investigation of the monthly mean values of the aerosol optical thickness carried out on the Baltic Sea coast at Norrköping (Persson 1999) in the years 1977–1997 indicates that much higher values may be expected in spring/summer than in autumn/winter.

The above results encouraged us to investigate the seasonal variability of this optical parameter in detail. Firstly, the frequency of occurrence of the aerosol optical thickness (at $\lambda = 555\,\mathrm{nm}$) for spring (March, April, May), summer (June, July, August) and autumn (September and November) is analysed for both daily and hourly means. The results are presented in Figs. 2a–c. They indicate that the most probable value of the aerosol optical thickness ranges between 0.05 and 0.1 for spring and 0.15–0.20 for summer conditions. These results are similar for the hourly and daily mean values of the aerosol optical thickness. For autumn, the daily mean aerosol optical thicknesses lie within the 0.1–0.15 range, but the peak is less sharp (see Fig. 2c) than in the case of the spring or summer values (Figs. 2a,b). Moreover, analysis of the frequency of occurrence based on the hourly mean values (Fig. 2) indicates that in autumn different values of the aerosol optical thickness occur with almost equal probability.

To examine the influence of advection of different air masses on the aerosol optical thickness, all data were separated into two classes, corresponding to two wind direction sectors: northern 270°–N–90° and southern 90°–S–270°. From the north, polar and arctic air masses, both continental and maritime, flow in over the Baltic Sea, whereas in the case of winds from the southern sector polar and tropical air masses of continental and maritime origin are expected. Moreover, in the case of winds from the northern sector, air masses are more likely to have spent a long time over the Baltic Sea before they reach its southern part. For each wind sector and each season, the mean value of the aerosol optical thickness and the Ångström parameters were calculated. Figs. 3a–c presents the seasonal variations in the frequency of occurrence of the aerosol optical thickness for two wind direction sectors. The wind direction frequency distributions during the measurements are also given.

The results presented in Table 4 indicate that regardless of the season the mean value of the aerosol optical thickness at 555 nm reaches its highest value for the southerly winds. The respective differences for spring, summer and autumn between the mean values for the southern and northern sectors are 0.046, 0.039 and 0.093. For averaged aerosol optical thickness spectra, the values of the Ångström parameter α are more similar to

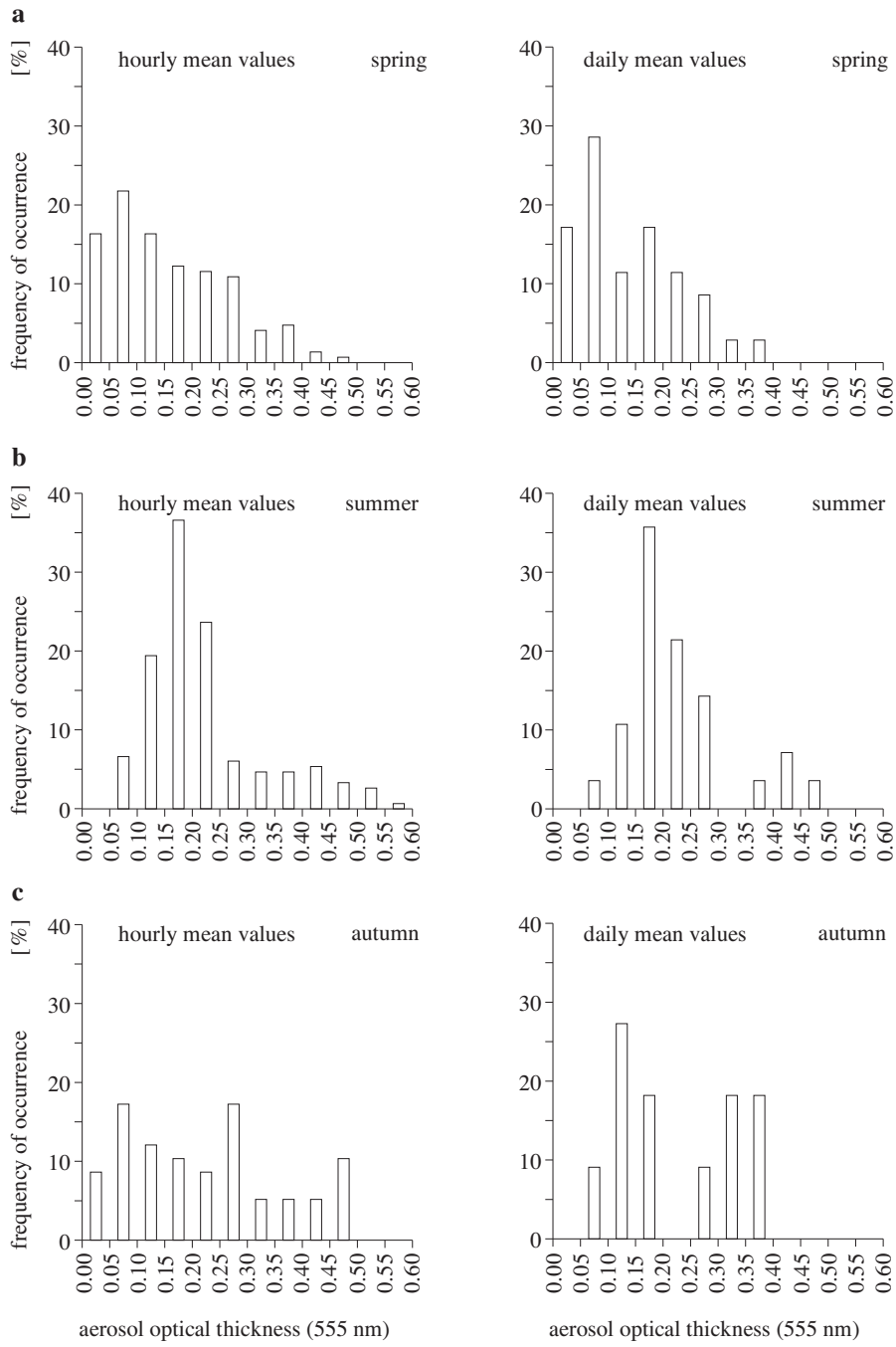
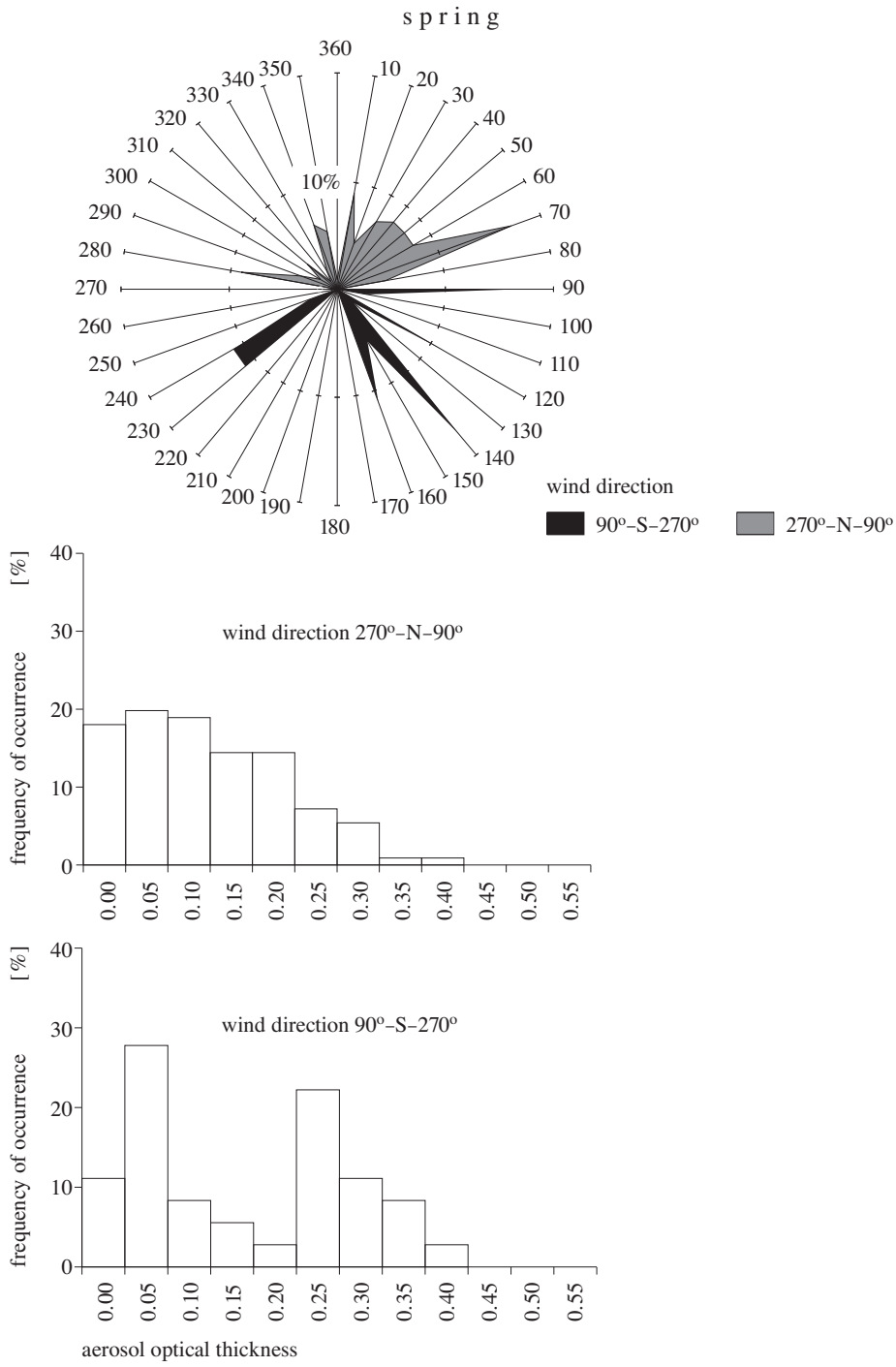


Fig. 2. Seasonal frequency of occurrence of the aerosol optical thickness at $\lambda = 555\,\mathrm{nm}$ for spring (March, April, May) (a), summer (June, July, August) (b), autumn (September and November) (c), calculated for the hourly and daily mean values. The sum of all frequencies is equal to 100% for each season

a


spring
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30
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50
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70
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240
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260
270
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290
300
310
320
330
340
350
10%
wind direction
90°–S–270°
270°–N–90°
[%]
frequency of occurrence
wind direction 270°–N–90°
wind direction 90°–S–270°
0.00
0.05
0.10
0.15
0.20
0.25
0.30
0.35
0.40
0.45
0.50
0.55
aerosol optical thickness

Fig. 3.

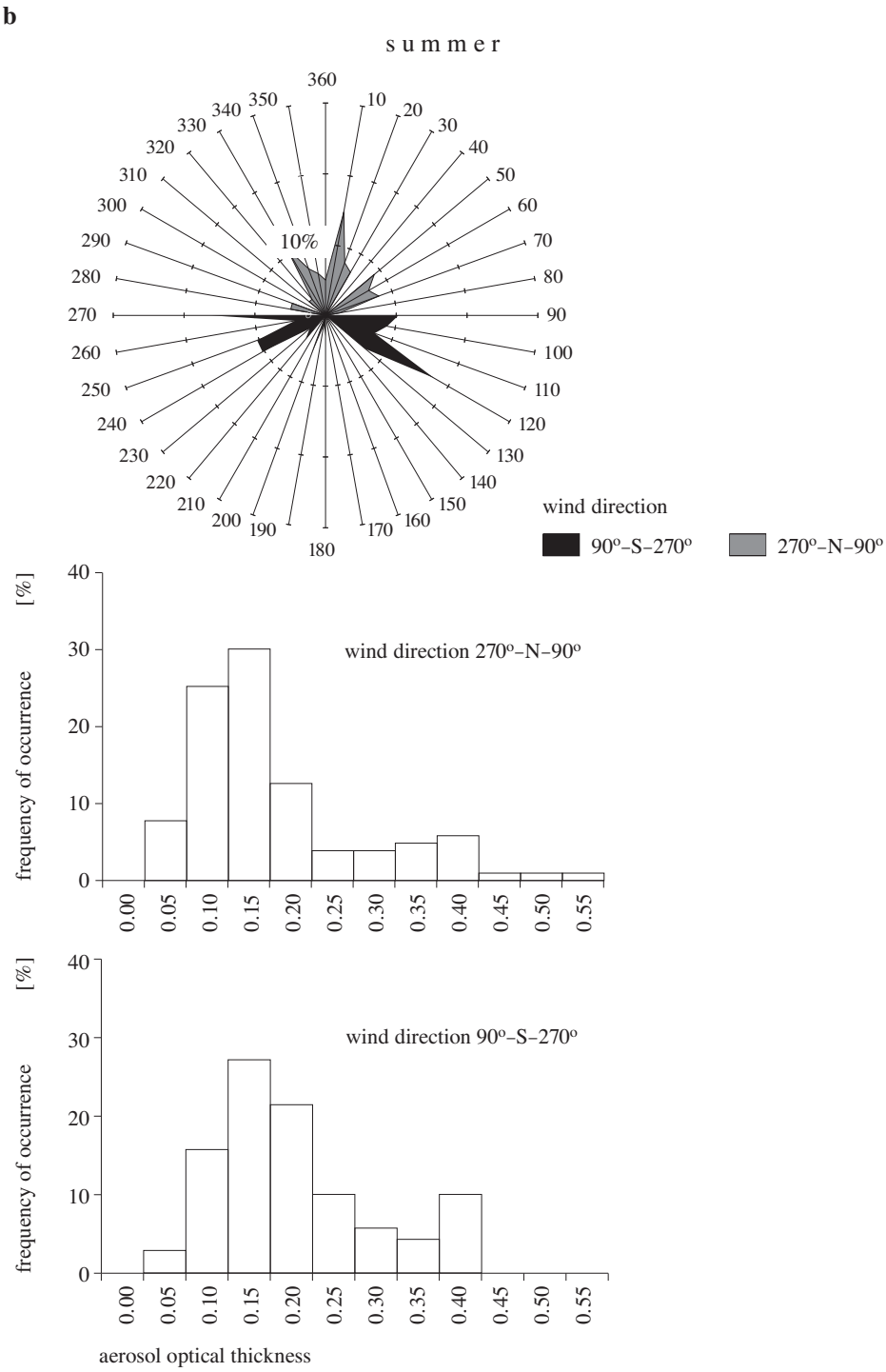

Fig. 3. (continued)

c

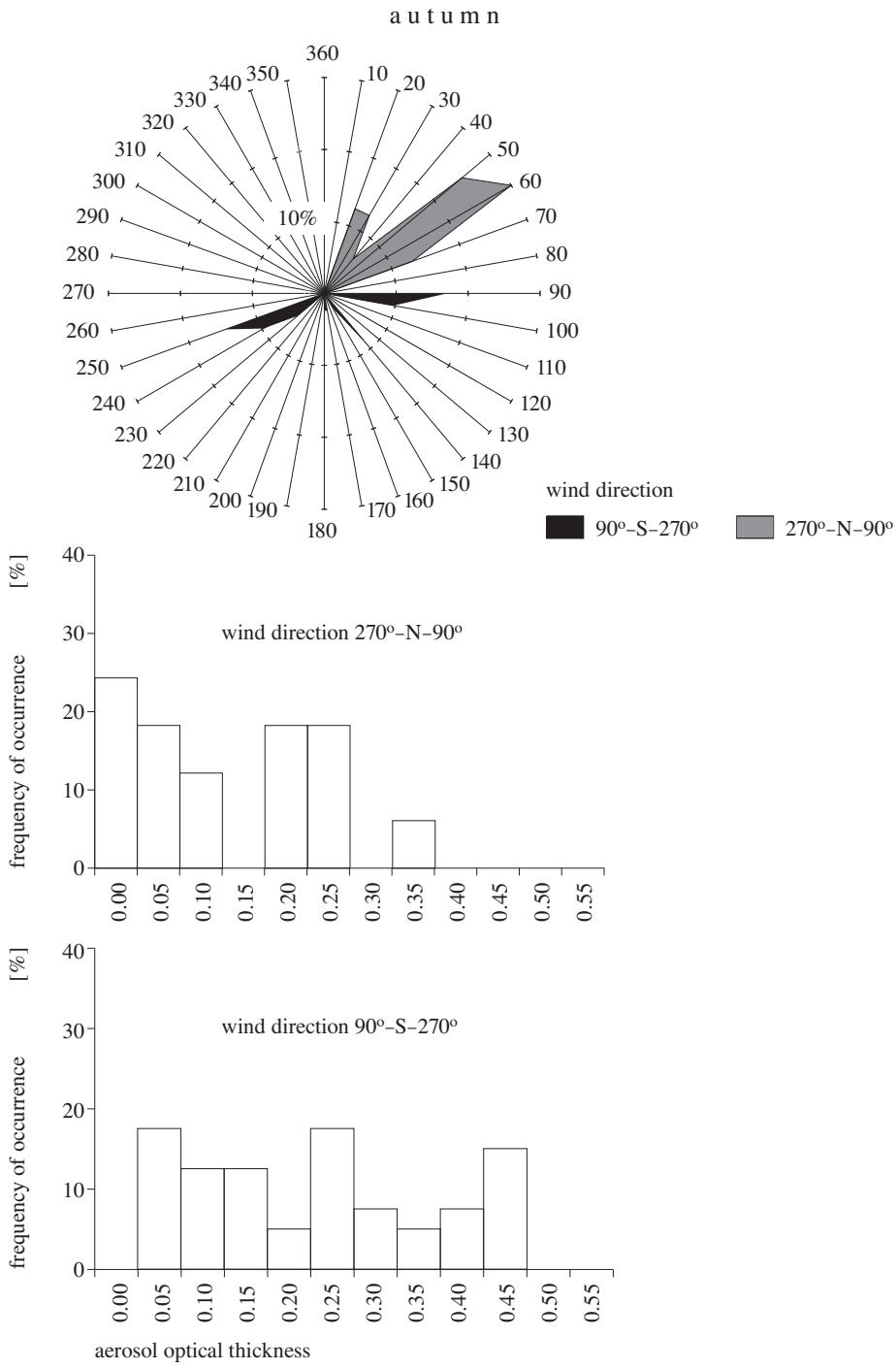

Fig. 3. (continued)

Fig. 3. Wind direction frequency distributions during the measurements and seasonal variation in frequency of occurrence of the aerosol optical thickness at $\lambda = 555$ nm, for two wind direction sectors – northern 270°–N–90° (polar and arctic air masses) and southern 90°–S–270° (polar and tropical air masses): spring (March, April, May) (a), summer (June, July, August) (b), autumn (September and November) (c). The sum of all frequencies is equal to 100% for each season

Table 4. Seasonal variability of the aerosol optical thickness and the Ångström parameters for λ in the 412 nm to 865 nm range

Season, wind direction	N	$< \tau_A(555) >$	σ_τ	$< \tau_A(\lambda) >$	
				γ	α
spring (March, April, May)	147	0.155	0.107	0.093	0.912
270°–N–90°	111	0.143	0.100	0.086	0.912
90°–S–270°	36	0.189	0.122	0.111	0.913
summer (June, July, August)	171	0.225	0.113	0.131	0.955
270°–N–90°	107	0.211	0.107	0.115	1.054
90°–S–270°	64	0.250	0.118	0.158	0.816
autumn (September, November)	58	0.225	0.138	0.124	1.041
270°–N–90°	16	0.158	0.113	0.099	0.822
90°–S–270°	42	0.251	0.139	0.134	1.095

N – the number of cases,

γ, α – were calculated on the basis of eq. (7) for the spectral values of the aerosol optical thickness averaged over a season.

those for continental aerosol type (Table 1) or mixed continental-maritime (*cf.* Table 1) than the maritime aerosol type, and varies in the 0.8–1.1 range for all the seasons and wind directions analysed.

The influence of the relative humidity on the aerosol optical thickness has previously been observed by many researchers, for example Covert *et al.* (1972), Gong *et. al.* (1987). In this paper this influence is also investigated. Cases where the wind speed did not exceed 6 m s^{-1} were used in the analysis. Whitecaps are scarce when wind speeds are below 6 m s^{-1}, so breaking waves are a rather inefficient local aerosol source in our data set. The results (Figs. 4a,b) show that an increase in the relative humidity results in an increase in the probability that high optical thickness values will occur. However, the relative humidity does not seem to influence the lower limit of the distributions (*e.g.* 10$^{\text{th}}$ percentiles). The mean aerosol optical thickness increases from 0.10–0.14 and from 0.07–0.15 for relative humidities of $< 70\%$ and winds from the southern and northern sector respectively to 0.23–0.29 and 0.16–0.21 for relative humidities of $> 70\%$.

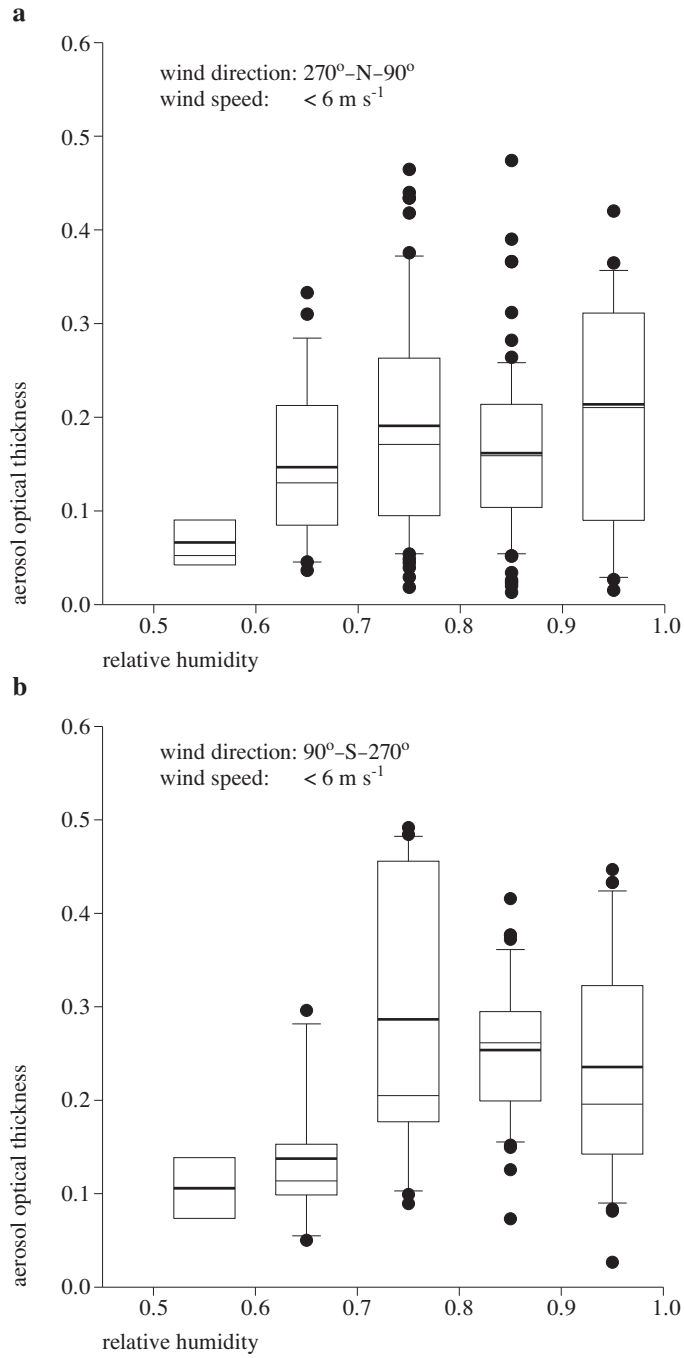

Fig. 4. The mean (thick line), median (thin line), 10^{th}, 25^{th}, 75^{th} and 90^{th} percentiles of the frequency distributions of the aerosol optical thickness at $\lambda = 555\,\text{nm}$ versus relative humidity for wind speeds less than $6\,\text{m s}^{-1}$ and two wind direction sectors: northern 270°–N–90° (a), southern 90°–S–270° (b)

5. Conclusions

- The lowest mean aerosol optical thicknesses at 555 nm 0.09–0.11 were recorded in March, April and November. The values obtained for June, July, August and September are comparable and are almost twice as high as those for March, April and November.
- In the southern Baltic area the mean aerosol optical thickness at 555 nm reaches its highest value when winds are from the southern sector, regardless of season. The mean optical thickness for winds from the directions 270°–90° (northerly directions) is 0.14 ($\pm$ 0.10) for spring, 0.21 ($\pm$ 0.11) for summer and 0.16 ($\pm$ 0.11) for autumn, whereas for winds from the directions 90°–270° (southerly directions) is 0.19 ($\pm$ 0.12) for spring, 0.25 ($\pm$ 0.12) for summer and 0.25 ($\pm$ 0.14) for autumn (The standard deviation of an individual measurement from the mean is given in parentheses).
- The values of the Ångström parameter α for the averaged aerosol thickness spectrum vary between 0.8 and 1.1, which is much higher than the typical values for 'pure' maritime aerosols – 0.4–0.5.
- An increase in relative humidity results in an increase in the probability of occurrence of high optical thicknesses. The variations in the relative humidity do not affect the expected minimum optical thickness values (the 10^{th} percentile).
- Both the mean and median of the optical thickness probability distributions increase with increasing relative humidity.

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