Aerosol optical and microphysical properties over the Atlantic Ocean during the 19th cruise of the Research Vessel *Akademik Sergey Vavilov*

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[1] This paper presents aerosol optical depths in the total atmospheric column, aerosol size distributions, number concentrations and black carbon mass concentrations at the deck level measured in October–December 2004 on board the R/V *Akademik Sergey Vavilov*. Aerosol optical depths measured within the spectral range 0.34–4.0 μm were close to background oceanic conditions (~0.04–0.08) in the high-latitude southern Atlantic. Angstrom parameters derived within 440–870 nm and 870–2150 nm spectral ranges did not exceed 0.6, yielding averages of 0.34 and 0.12, respectively. The mass concentration of black carbon varied within the range 0.02–0.08 μg/m^3 in the 34–55°S latitudinal belt. The average of 0.04 μg/m^3 (s.d. ~0.015) is close to the reported results for the remote areas of the South Indian Ocean. Aerosol volume size distributions measured within the size range of 0.4–10 μm can be characterized by a geometric volume mean radius ~3 μm. This is consistent with the columnar retrievals reported by the Aerosol Robotic Network (AERONET).

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1. Introduction

[2] The accuracy of the direct aerosol forcing computations depends mainly on the quality of aerosol models. The quality of aerosol models, in turn, strongly depends on the amount of empirical data and extensive coverage area. Collecting data over the oceans has never been an easy task. Being time, and labor consuming and also very expensive, those measurements made from research vessels were rare and definitely did not cover all regions of the World Ocean. Measurements in coastal zones and from island sites made within the framework of the internationally federated AERONET program [*Holben et al.*, 1998] or made by individual institutions do not fully solve the problem of disparate data coverage. There are still many areas over the oceans where information on atmospheric optical properties is completely or partly missing. In some areas ship-based measurements is the only data collection option.

[3] Various reviews published to date on aerosol optical depth measurements over the oceans [*Barteneva et al.*, 1991; *Smirnov et al.*, 2002; *Sakerin and Kabanov*, 2002; *Quinn and Bates*, 2005] illustrate that Sun photometer data were taken more frequently over the northern and central Atlantic, including inland seas and coastal areas, the Central Pacific, and to some extent over the northern Indian Ocean. The atmosphere of the southern Atlantic, especially south of 40° ("roaring forties") lacks not only reliable data but in point of fact any data on aerosol optical depth and can be considered as one of the most poorly studied parts of the World Ocean.

[4] Because of its absorption properties and long residence time in the atmosphere black carbon is an important anthropogenic aerosol component [*Penner and Novakov*, 1996]. According to *Jacobson* [2001], black carbon's contribution to the direct radiative forcing exceeds that due to CH_4. The important role of black carbon in the atmospheric radiation balance has created a demand for more data acquisition and analysis [see, e.g., *Novakov et al.*, 1997, 2003, 2005; *Hansen et al.*, 2000]. Black carbon measurements are sparse over the oceans, especially in remote oceanic areas.

[5] Despite aerosol size distribution measurement results can be subject to various specific instrumental biases (see, e.g., comprehensive analysis by *Reid et al.* [2003, 2006]), we believe that new experimental data can still be a very useful addition, especially to such poorly studied areas as the southern Atlantic.

[6] In the current paper we report aerosol optical depth measurements over the Atlantic Ocean, aerosol size distribution and number concentration measurements at deck

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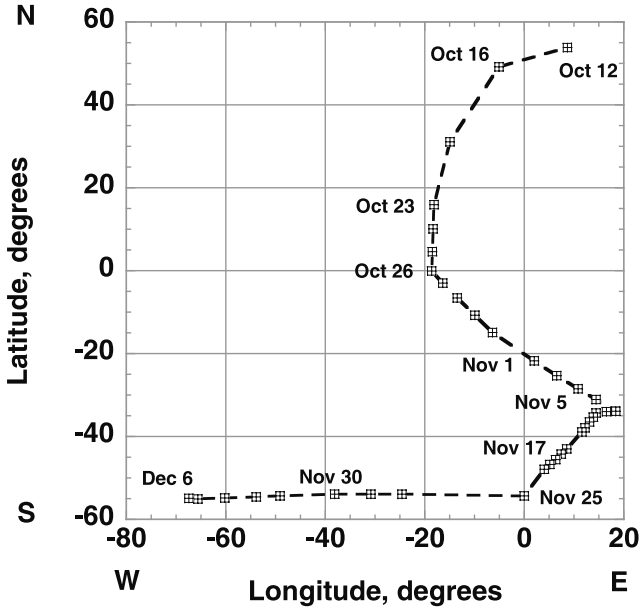

Figure 1. R/V *Akademik Sergey Vavilov* cruise track.

level, acquired within the 0.4–10 μm size range, and black carbon concentration data.

2. Instrumentation and Data Collection

[7] Aerosol measurements were made from October to December 2004 on board the R/V *Akademik Sergey Vavilov*. The cruise area included a transect in the Atlantic from the North Sea to Cape Town, South Africa and then a crossing in the South Atlantic to Ushuaia, Terra del Fuego, Argentina (Figure 1). Aerosol measurements included (1) columnar aerosol optical depth using the automated Sun photometer SP-5 [*Sakerin and Kabanov*, 2002] and a hand-held Sun photometer Microtops II [*Morys et al.*, 2001]; (2) deck-level aerosol size distribution measurements acquired within the 0.4–10 μm diameter range using the electric particle counter AZ-5 [*Sokolov and Sergeyev*, 1970]; and (3) black carbon mass concentration measurements using an in-house soot measuring device (aethalometer) [*Kozlov et al.*, 1997].

[8] Instruments were collocated on the upper deck of the ship in order to avoid Sun photometer line-of-sight obstructions and to minimize the influence of local aerosol sources.

[9] The SP-5 multiwavelength Sun photometer is an automated device specifically designed to measure columnar optical depth and water vapor content as described in detail by *Sakerin and Kabanov* [2002] and *Sakerin et al.* [2005]. In the UV spectral range the SP-5 has a SiC detector for two spectral channels, a silicon photodiode is used for 10 visible and near-IR channels, and a pyroelectric detector is deployed for the short-wave IR channels. Basic spectral characteristics of the interference filters are summarized in Table 1. The photometer is mounted on a two coordinate (zenith/azimuth) turntable. Electric drives are controlled by a system of photosensors, including four photodiodes for coarse pointing and a four-sector photodiode for precise tracking.

[10] The measurements were carried out in weather conditions when the solar disk was free of clouds and solar zenith angle was less than 80°. The instrument was precalibrated using the Langley method but after some precruise repairs the calibration was repeated while at sea on clear days during the morning hours. The calibration scheme and the procedure for computing optical depth and columnar water vapor content (in cm of precipitable water) are given by *Sakerin and Kabanov* [2002] and *Kabanov and Sakerin* [1997]. The principal idea of the computational algorithm is to take into account molecular scattering and gas absorption at the initial stage, dividing the measured signal by the transmission functions computed using the LOWTRAN-7 spectroscopy [*Kneizys et al.*, 1988] and Sun photometer spectral functions. The uncertainty of aerosol optical depth in the UV channels does not exceed 0.02, being between 0.01 and 0.02 for visible and near IR range, and increasing to 0.02–0.03 for the SWIR channels.

[11] We averaged a 30-min measurement period into one data point. Daily averages were calculated as a simple arithmetic mean of the 30-min averages. We characterized the temporal and latitudinal distribution of aerosol optical depth using the daily averaged values and Angstrom parameter α computed as a square-linear fit to the classical equation $\tau_a \sim \lambda^{-\alpha}$. The Angstrom parameter α was derived for two spectral ranges: 440–870 nm and 870–1250 nm. The parameter from the first spectral range, α_{vis} provides a common basis for comparison with previously reported results and the second (α_{nir}) can be used for aerosol characterization in the SWIR spectral range, and as an indication of the extent to which the variability of microstructure affecting one spectral range (vis/nir) is dependent on the microstructure variability affecting a second spectral range (nir/swir) [*Villevalde et al.*, 1994; *Smirnov et al.*, 2003a].

[12] Data acquired with the handheld Microtops II Sun photometer [*Morys et al.*, 2001] were reported elsewhere [*Smirnov et al.*, 2006]. Briefly, direct Sun measurements were taken in five spectral channels at 340, 440, 675, 870, and 940 nm. The instrument was calibrated at the NASA Goddard Space Flight Center against the AERONET reference CIMEL Sun/sky radiometer [*Holben et al.*, 1998, 2001]. The estimated uncertainty of the optical depth in each channel did not exceed plus or minus 0.02. The number of measurements averaged into one data point (a series) was not less than 5 during a 3-min period. The number of series during the day varied from 1 to 33. Aerosol

Table 1. Filter Characteristics of SP-5 Sun Photometer

	UV Range	VIS and NIR Range	SWIR Range
Field of view, deg	0.92	1.50	1.15
Number of channels	2	10	4
Filter wavelengths, nm	340, 370	420, 440, 480, 550, 630, 680, 780, 870, 940, 1060	1240, 1560, 2150, 4000
FWHM, nm	5	5–15	10–50
Time of one cycle, s	50	50	50

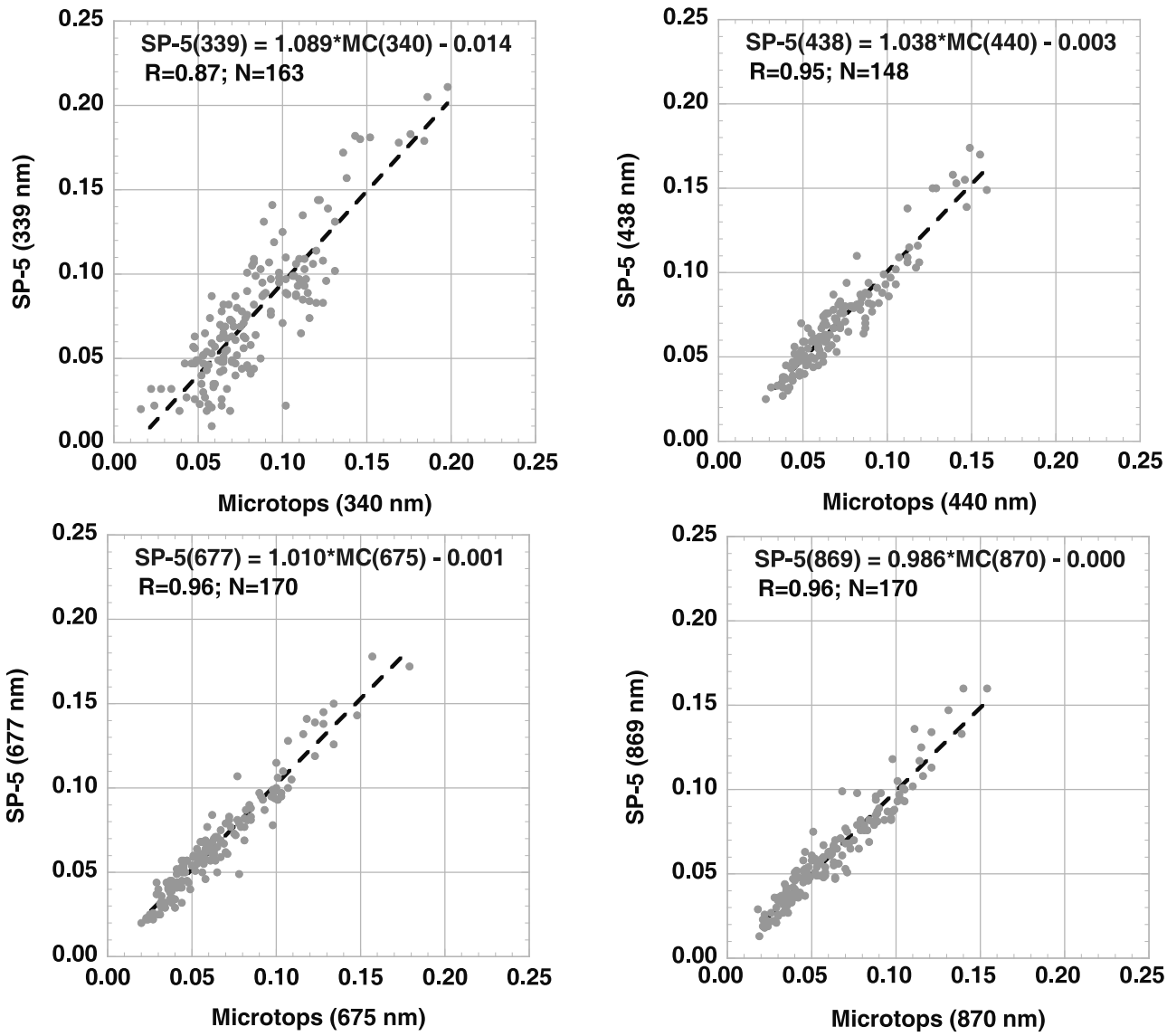

Figure 2. Sun photometer data comparison (SP-5 versus Microtops).

optical depth was retrieved by applying the AERONET processing algorithm (Version 2) to raw data. Details of the processing algorithm are available at http://aeronet.gsfc.nasa.gov and partly in the work by *Smirnov et al.* [2004]. In the current work we will use Microtops data only for comparison.

[13] To measure aerosol size distributions over the oceans the commercially available electric particle counter AZ-5 [*Sokolov and Sergeyev*, 1970] was modernized. A spectra analyzer was built in, and the measurement process and registration were fully automated and computerized. A simple bulb (white light) was used as a source and a photomultiplier was used as a detector. The scattered beam intensity is registered at 90° scattering angle. The working volume is 0.6 mm^3. The instrument has 12 size channels covering the size range from 0.4 through 10 μm. A similar optical scheme was used in the particle counters produced by Royco Instr. [*Lui et al.*, 1974]. Absolute calibration of the optical particle counter AZ-5 was provided by the manufacturer.

[14] The measurement principle of the built in-house soot measuring device (aethalometer) [*Kozlov et al.*, 1997] is similar to that employed in the aethalometer designed by *Hansen et al.* [1984]. The instrument continuously measures diffuse light attenuation by layers of particles while they keep accumulating on the filter surface. The registered signal is proportional to the particle-surface-area concentration of soot and thus its mass concentration in the air. The method robustness was proven by *Rosen and Novakov* [1983] and *Clarke* [1982] among many others. The theoretical rationale behind the Hansen method is that diffuse light attenuation by the aerosol particles on a filter depends mainly on aerosol absorption not scattering properties.

[15] In our instrument the airflow moves through a hose with a diameter ~8 mm and 2 m length before it reaches the working volume with the filter. Coarse mode aerosols with diameters >1 μm precipitate along the way. Thus the aethalometer registers soot only for fine mode aerosol particles. The flow rate is ~5 L/min.

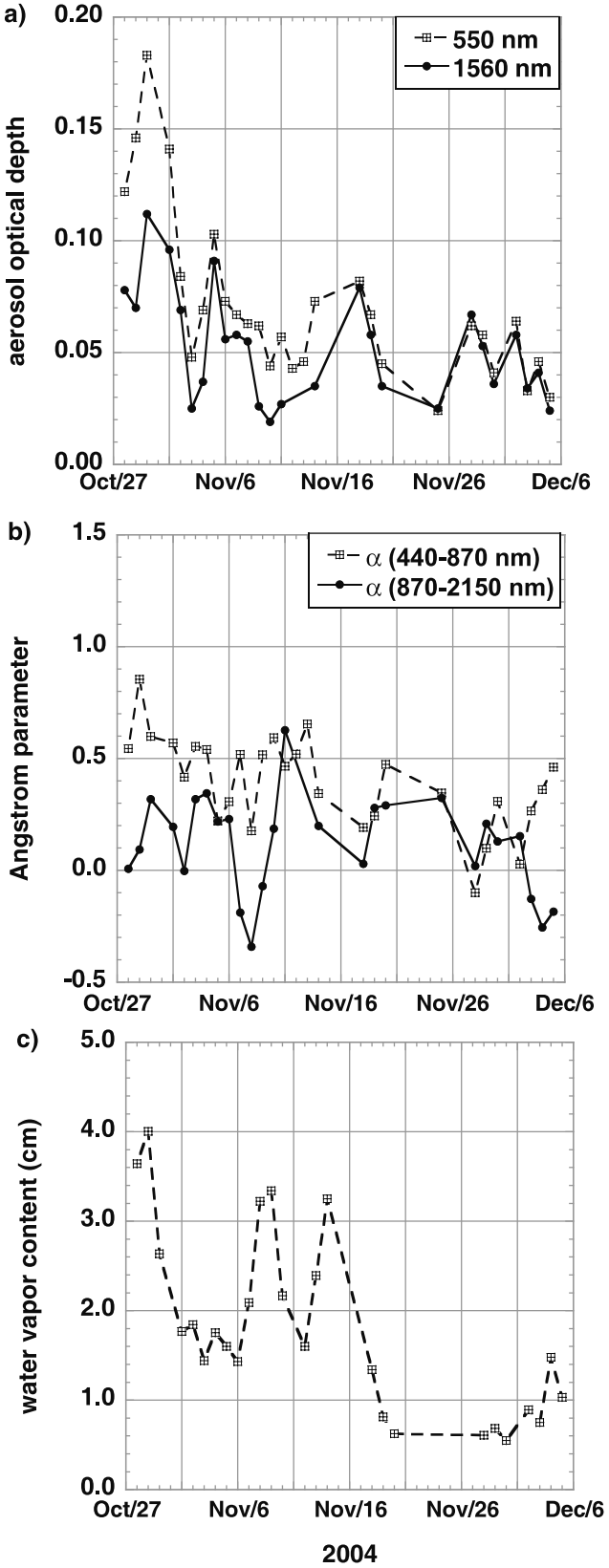

Figure 3. Temporal distribution of daily averaged (a) aerosol optical depth at wavelengths 550 and 1560 nm, (b) Angstrom parameter computed within 440–870 and 870–2150 nm spectral range, and (c) columnar water vapor content.

[16] We conducted an absolute calibration of the aethalometer in the laboratory, comparing optical and gravimetric measurements [*Baklanov et al.*, 1998]. The soot particles used in the process range between 50 and 200 nm. The minimum registered mass soot concentration was ~0.02 μg/m^3 for a pumped flow volume of ~35 L. Such sensitivity will guarantee reliable results even in the background conditions with very low aerosol amounts.

3. Results

3.1. Sun Photometer Data Comparison

[17] The uncertainty in the aerosol optical depth measurement depends on various factors: calibration uncertainty, filter degradation, scattered light within the instrument field of view, and computational uncertainties due to uncertainties in molecular scattering and gaseous absorption. Additional sources of errors at sea include ship motion and obstruction of the Sun by masts and antennas. We compared aerosol optical depths measured by the SP-5 and Microtops II which had different calibration and processing procedures. Also note that the different shape of filter functions for two instruments might also contribute to differences in τ_a. For comparison we used optical depths from both Sun photometers taken within 6 min from each other.

[18] Figure 2 presents aerosol optical depth regressions for common channels around 340, 440, 675 and 870 nm. One can observe that the correlation coefficients between the two data sets are quite high, intercepts are close to zero, and systematic error does not exceed 9%. For the optical depth range considered (see Figure 2) this error could be treated as insignificant. Absolute differences in general do not exceed 0.02, being slightly higher (~0.03) for a number of instances at a wavelength of 340 nm. Columnar water vapor contents measured by both instruments were consistent also.

[19] Therefore we can conclude that aerosol optical depths measured by two independently calibrated and processed systems proved to be reliable and comparable.

3.2. Aerosol Optical Parameters Over Areas in the Southern Atlantic

[20] Data acquisition using the SP-5 was started on 28 October when the ship already reached latitude 7°S. Daily averages of aerosol optical depth at 550 and 1560 nm wavelength, α_{vis} and α_{nir}, and columnar water vapor content are shown in Figures 3a–3c. Figure 3a shows good correlation between optical depth variations in the visible and SWIR channels. From the middle of November through to the end of the cruise almost no difference was observed between τ_a (550 nm) and τ_a (1560 nm). This is an indication of coarse mode aerosol particles playing a major role in the aerosol optical depth spectral behavior. The difference and lack of evident correlation between α_{vis} and α_{nir} (Figure 3b) illustrates the fact that the coarse mode of the aerosol size distribution varies, to a certain extent, independently of the fine mode (confirmed by very low correlation coefficient ~0.15 between α_{vis} and α_{nir}). Water vapor content (or columnar precipitable water amount) ranged from 0.5 to 4 cm. This variation was more regional-dependent and thus was more characteristic of a synoptic pattern. Minimal values were observed at latitudes south of

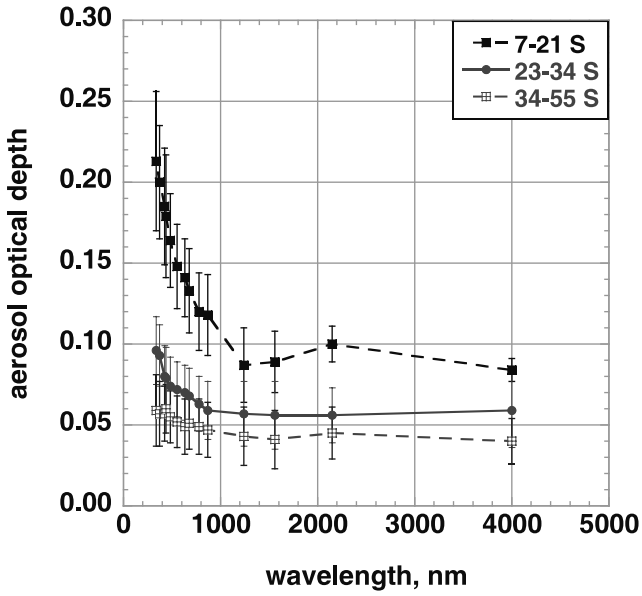

Figure 4. Mean regional aerosol optical depths. The vertical bars indicate plus or minus one standard deviation.

40° while maximum water vapor content was measured near the equator at the beginning of the measurement period (Figure 3c).

[21] Our optical depth measurements were grouped into three subsets (Figure 4). The grouping was selected in order to present the variability of our optical data in a manner which was similar to *Voss et al.* [2001]. The optical depth in the first grouping of Figure 4 (the South Atlantic Tropical region 7–21°S, 4 days of measurements) was higher than during the Aerosols99 Experiment; however, *Voss et al.* [2001] pointed out a notable latitudinal variability of optical depth in this area. Ship-based optical depth measurements by *Voss et al.* [2001] did suggest certain aerosol stratification with moderate aerosol amount at the upper levels. This was confirmed by lidar measurements, although the columnar aerosol amount was not high (τ_a (550 nm)$\sim$0.10). The AERONET data from the Ascension Island suggest [*Holben et al.*, 2001; *Smirnov et al.*, 2002] that on a number of occasions dust and biomass burning aerosol can be transported from Africa and influence optical properties over the ocean. Statistical characteristics of the aerosol optical parameters (τ_a (500 nm)$\sim$0.15, $\alpha_{\rm vis} \sim 0.64$) are close to those reported for this region by *Barteneva et al.* [1991], *Sakerin and Kabanov* [1999], and *Smirnov et al.* [2002]. The aerosol optical depth in the South Atlantic subtropical marine area (25–34°S, 7 days of measurements, including 4 full days in the port of Cape Town) was low (τ_a (550 nm) $\sim$0.07, $\alpha_{\rm vis} \sim 0.40$), because of the easterly winds at various pressure levels. Back trajectory analysis indicated aerosol sources to be over the Atlantic Ocean at the pressure levels considered (between 1000 and 500 mbar). Optical properties in the South Atlantic subtropical marine area agree very well with earlier studies by *Volgin et al.* [1988], *Voss et al.* [2001], *Smirnov et al.* [2002], and *Knobelspiesse et al.* [2004].

[22] The third set was taken between 9 November and 5 December when the ship was moving south from latitude 34°S to 55°S (17 measurement days). The aerosol optical depth at 550 nm in that region varied between 0.02 and 0.08 and the average (τ_a (550 nm)$\sim$0.05 associated with an $\alpha_{\rm vis} \sim 0.34$) was lower than in the Pacific Ocean as reported by *Volgin et al.* [1988], *Villevalde et al.* [1994], and *Smirnov et al.* [2002, 2003b].

[23] The wind speed range for the area south of 40° latitude was 5–15 m/s during the measurement period. We did not find any correlation between wind speed and instantaneous aerosol optical depths. Even considering only measurements taken in the transparent atmosphere of the southern Atlantic we did not find any noticeable trend. In this regard we would like to point out again the necessity of considering averaged wind speed over a previous time period regardless if we analyze the near surface or columnar extinction measurements [*Gathman*, 1983; *Hoppel et al.*, 1990; *Flamant et al.*, 1998; *Smirnov et al.*, 2003b]. However, such detailed wind speed information was not available to us. The influence of the surface level wind speed on the columnar aerosol optical parameters is a difficult problem. The link is not easy to detect given the many masking effects which contribute to the atmospheric optical state (e.g., relative humidity, background aerosol, aerosol aloft, etc.).

[24] The aerosol optical depth in the short-wave IR range had almost neutral spectral dependence. The Angstrom parameter $\alpha_{\rm nir}$ did not change much being on average $\sim$0.10 for the three subsets considered above (Figure 4). Shown standard deviations of τ_a in the SWIR are either smaller than in the visible (for measurements between 7–21°S) or about the same (from 34°S to 55°S). The few publications which exist [*Wolgin et al.*, 1991; *Villevalde et al.*, 1994; *Shiobara et al.*, 1996; *Vitale et al.*, 2000] showed both neutral and selective spectral τ_a dependence in the SWIR range.

[25] Measurements in the 1240–4000 nm spectral range permitted the partition of the optical depth into fine and coarse optical components. We define τ_a (coarse) as the average τ_a over the 1240–4000 nm spectral range (justified by the fact that optical depth spectral changes are small). Fine mode optical depth can be presented as the difference between τ_a (550 nm) – τ_a (coarse). Thus defined, the optical depth components were of comparable magnitude for the 7–21°S latitudinal belt (τ_a (fine)$\sim$0.06, τ_a (coarse)$\sim$0.09) while the coarse mode was dominant for the two other subsets presented in Figure 4 (τ_a (fine)$\sim$0.01, τ_a (coarse)$\sim$0.04–0.06).

[26] Figure 5 compares known reported results of aerosol optical depth measurements in the Southern Ocean with the current study. In general mean optical depths at a 550 nm wavelength are similar in magnitude while low $\alpha_{\rm vis}$ are also evident in the approximate neutrality of most of the spectra. To illustrate the difference with other areas in the Atlantic Ocean we show in Figure 5 our measurements [*Sakerin and Kabanov*, 2002] in the remote Atlantic not influenced by Saharan dust and data acquired in the North Atlantic between 64° and 66° N as reported by *Villevalde et al.* [1994]. It can be seen that τ_a (550 nm) and $\alpha_{\rm vis}$ are slightly higher for Central and North Atlantic than for the southern latitudes.

3.3. Soot and Aerosol Concentration Measurements

[27] Soot and aerosol concentration measurements started on 10 October and continued daily until 2 December. Black

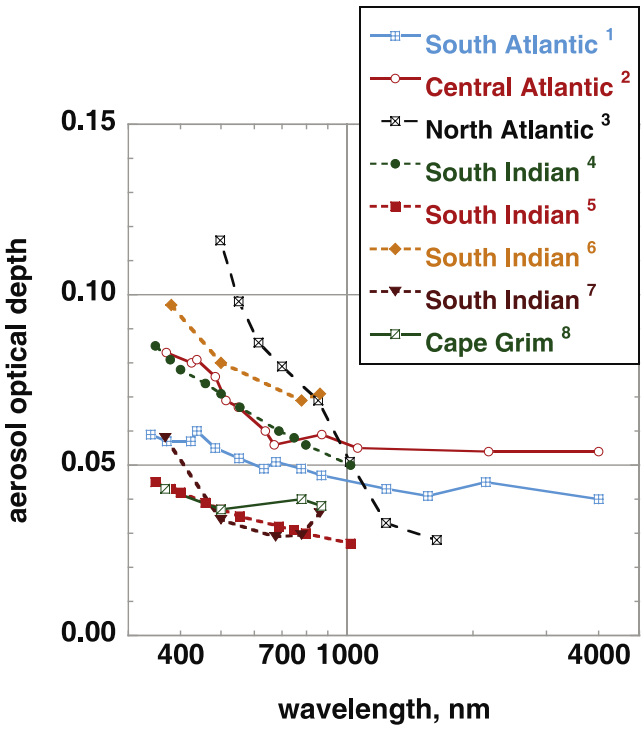

Figure 5. Aerosol optical depth spectra comparison for various oceanic areas: 1, South Atlantic Ocean, 34–55°S, current work; 2, central Atlantic Ocean, 35–50°N [*Sakerin and Kabanov*, 2002]; 3, North Atlantic Ocean, 64–66°N [*Villevalde et al.*, 1994]; 4, South Indian Ocean, 39–40°S, 1981 [*Barteneva et al.*, 1991]; 5, South Indian Ocean, 47–65°S, 1987 [*Barteneva et al.*, 1991]; 6, South Indian Ocean, 41–51°S, 1995, ACE-1 [*Quinn and Bates*, 2005]; 7, South Indian Ocean, 42–67°S, 1981 [*Matsubara et al.*, 1983]; 8, Cape Grim, 40°S [*Wilson and Forgan*, 2002].

carbon mass concentration daily averages, number concentration daily averages (within the 0.4–10 μm diameter range) and corresponding standard deviations are shown in Figure 6a. The latitudinal dependence is presented in Figure 6b while a partition into latitudinal belts is displayed in Figure 6c.

[28] Figures 6a and 6b show some decrease of the number concentrations (approximately from 6.5 to 3.5 cm^{-3}) from the equator to Cape Town (34°S). The total number density (N) reached a minimum south of Cape Town and, after varying around a mean level of ~5 cm^{-3} (from 11 through 24 November), N decreased again to ~2–3 cm^{-3} in the area to the south of 52°. The latitudinal dependence of N_1 ($0.2 \leq r \leq 0.5$ μm) and N_2 ($0.5 \leq r \leq 5$ μm) reveals, overall, no increase of large particle fraction in the areas south of 34°, despite a general increase in wind speed. Generally speaking the variability of aerosol concentration can be quite complex. Contrary to soot, which is transported mainly from the continents, the sea surface itself is quite a strong source of aerosol particles [see, e.g., *O'Dowd et al.*, 1997; *Quinn et al.*, 1998; *Glantz et al.*, 2004], producing aerosols within (but not limited to) the radius range of our photoelectric counter ($0.2 \leq r \leq 5.0$ μm). Therefore the number density of aerosol particles depends in a complicated manner on the transport

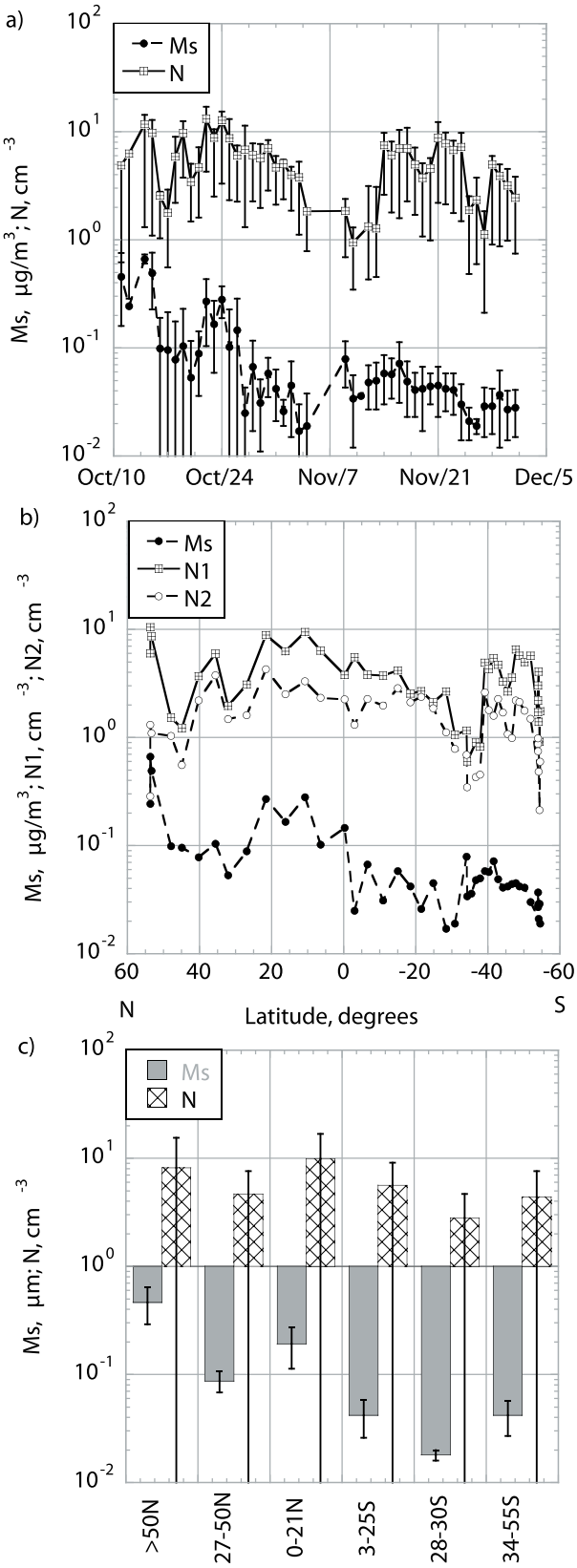

Figure 6. (a) Temporal, (b) latitudinal, and (c) regional distributions of the daily averaged black carbon mass concentrations (M_s), aerosol number concentrations within the radii range 0.2–5 μm (N), 0.2–0.5 μm (N_1), and 0.5–5 μm (N_2). The vertical bars indicate plus or minus one standard deviation.

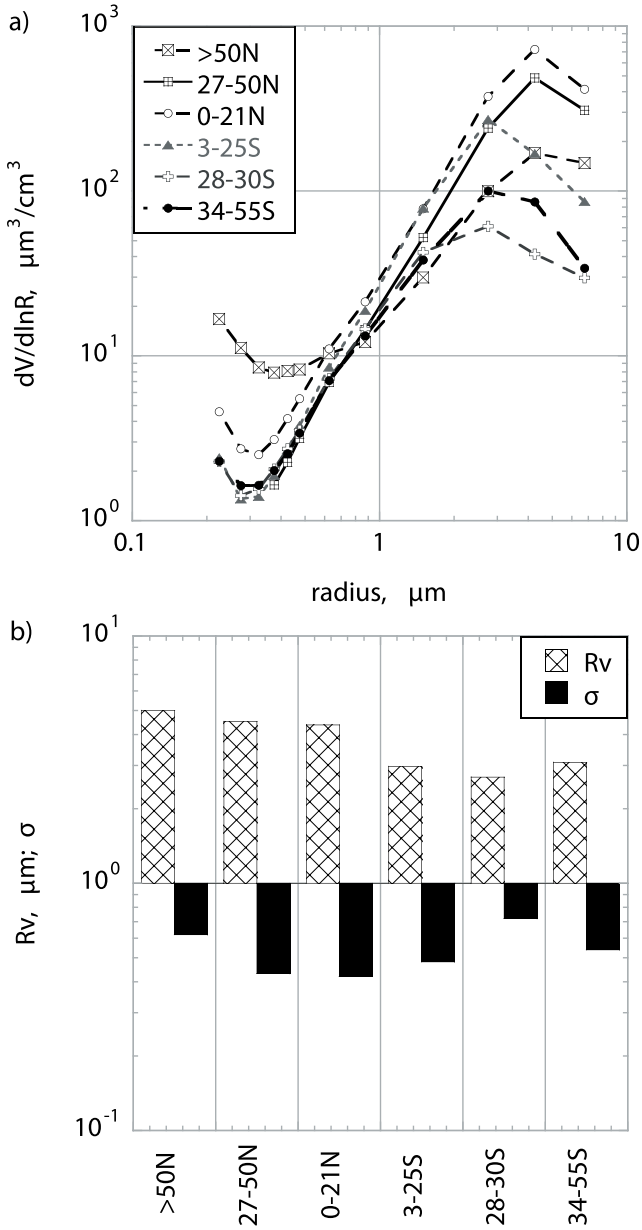

Figure 7. (a) Average aerosol volume size distributions and (b) geometric mean radii and geometric standard deviations for various regions (latitudinal belts).

from continents, on the state of the sea surface, and on the radius range considered.

[29] The BC mass concentration decreases with latitude (Figures 6a and 6b) on average, from 0.06 μg/m^3 at the equator to ~0.03 μg/m^3 at the latitudes greater than 50°S. The lowest concentrations were measured around 30°S, when the ship was approaching the Cape Town area, and in the southern Atlantic at ~55°S. This is consistent with the results reported by *Moorthy et al.* [2005] for the Indian Ocean and the Southern Ocean, where the BC mass decreased in the southern hemisphere from 0.3 μg/m^3 in the equatorial zone to less than 0.05 μg/m^3 in the Indian sector of the Southern Ocean (up to 56°S). Division into latitudinal districts (Figure 6c) emphasizes the low concentration of black carbon in the Southern Hemisphere to the south of 3°.

[30] Aerosol volume size distributions which have been temporally averaged over a day and then spatially averaged over the latitudinal belts are presented in Figure 7a. The mode radii of size distributions measured north of the equator (ranging between 4.3 and 5.0 μm) are bigger than those obtained to the south of the equator (ranging from 2.7 to 3.1 μm). The number of averaged days varied from 2 for the 28–30°S belt and 22 for the 34–55°S region. The volume geometric mean radii and geometric standard deviations are shown in Figure 7b. The numbers are consistent with the review paper by *Smirnov et al.* [2002] and the maritime component model based on the AERONET data [*Smirnov et al.*, 2003a]. The geometric mean radius for the most statistically representative subset (34–55°S) is slightly higher that recommended in the review paper by *Reid et al.* [2006].

4. Conclusions

[31] A summary of the optical parameters, microphysical parameters, and soot concentrations for the less frequently studied Southern Atlantic region is presented in Table 2.

[32] Table 2 shows that atmospheric aerosol optical parameters (τ_a(500 nm) ~0.05 and α_{vis} ~ 0.34) in the southern Atlantic between 34°S and 55°S are similar to typical values measured in other remote oceanic areas. However, the employment of a wider spectral range in the current study ensures that this data is particularly unique and valuable. In the SWIR spectral range the wavelength dependence of aerosol optical depth remains almost neutral (α_{nir} ~ 0.12).

[33] The BC mass concentrations in the Southern Hemisphere were low (~0.04 μg/m^3) and relatively stable.

[34] The volume geometric mean radius R_v ~ 3μm and the geometric standard deviation σ ~ 0.6 of the volume size distribution derived from in situ aerosol measurements are consistent with the parameters derived from AERONET data [*Smirnov et al.*, 2003a].

[35] **Acknowledgments.** The authors wish to acknowledge the financial support of the Russian Academy of Sciences through the Program of Fundamental Studies within the framework of the Project "Investigation of

Table 2. Statistical Characteristics and Variability of the Columnar Aerosol Optical Parameters and Aerosol Microphysical Characteristics at the Deck Level Measured in the Southern Atlantic Between 34 and 55°S

	τ_{a550}	τ_{a1560}	α_{vis}	α_{nir}	M_s, μg/m^3	N, cm^{-3}	N_2, cm^{-3}	R_v, μ	σ
Mean	0.05	0.04	0.34	0.12	0.042	4.39	1.20	2.97	0.56
s.d.	0.02	0;02	0.20	0.23	0.015	2.54	0.71	0.38	0.07
Minimum	0.02	0.02	−0.10	−0.26	0.019	0.95	0.21	2.17	0.45
Maximum	0.08	0.08	0.66	0.63	0.079	8.75	2.61	3.68	0.74

spatial and temporal variability of aerosol components over the oceans." We would like to extend our thanks to the crew of the R/V *Akademik Sergey Vavilov.* The authors (B. N. Holben and A. Smirnov) thank Hal Maring (NASA Headquarters) and Michael King (EOS Project Science Office) for their support of AERONET. We thank Norman T. O'Neill at University of Sherbrooke for his comments and suggestions, Valery S. Kozlov and Vladimir P. Shmargunov for technical support of the black carbon measuring instrumentation, and the anonymous reviewer for suggestions that improved the paper.

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