



1 **Radiative characteristics of aerosol under smoke mist conditions in Siberia**
2 **during summer 2012**

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13 **Abstract.** Microphysical and optical properties of aerosol were studied during mega-fire event in summer
14 2012 over Siberia using ground-based measurements of spectral solar radiation at AERONET site in Tomsk
15 and satellite observations. The data were analyzed using multiyear (2003-2013) measurements of aerosol
16 characteristics under background conditions and for less intense fires, differing in burning biomass type, stage
17 of fire, remoteness from observation site, etc. (“ordinary fires”). In June – August 2012, the average aerosol
18 optical depth (AOD, 500 nm) had been 0.95 ± 0.86 , about a factor of 6 larger than background values
19 (0.16 ± 0.08), and a factor of 2.5 larger than in “ordinary smokes. The AOD values were extremely high on July
20 24-28 and reached 3-5. Comparison with satellite observations showed that ground-based measurements in the
21 region of Tomsk not only reflect the local AOD features, but also are characteristic for the territory of the
22 Western Siberia as a whole. Single scattering albedo (SSA, 440 nm) in this period ranged from 0.91 to 0.99
23 with the average of ~ 0.96 in the entire wavelength range of 440-1020 nm. The increase in absorptance of
24 aerosol particles ($SSA(440\text{ nm})=0.92$) and decrease in SSA with wavelength, observed in “ordinary smokes”,
25 agree with the data of multiyear observations in analogous situations in boreal zone of USA and Canada.
26 Volume aerosol size distribution in smoke mist and ordinary smokes had bimodal character with significant
27 prevalence of fine mode particles, but in summer 2012 the mean median radius and the width of the fine mode
28 distribution somewhat increased. In contrast to data of multiyear observations, in summer 2012 an increase in
29 the volume concentration and median radius of the coarse mode was observed with the growing AOD.

30 The calculations of the “average” radiative effects of smoke and background aerosol are presented. As
31 compared to background conditions and “ordinary smokes”, under the conditions of smoke mist the cooling
32 effect of aerosol considerably intensifies: direct radiative effects (DRE) at the bottom (BOA) and at the top of
33 the atmosphere (TOA) are -13, -35, and - 60 W m⁻² and -5, -14, and -35 W m⁻² respectively. The maximal
34 values of DRE were observed on July 27 (AOD(500 nm)=3.5), when DRE(BOA) reached -180 W m⁻², while
35 DRE(TOA) and DRE of the atmosphere were -80 W m⁻².

36



37 *Key words:* Sun-sky radiometer, AERONET, smoke mist in Siberia 2012, optical and microphysical
38 properties of aerosol particles, diurnally radiative effects of smoke and background aerosol

39

40 **1 Introduction**

41

42 Massive forest and peat fires are the strongest source of aerosol-gas emissions on the territory of boreal zone
43 of the Northern Eurasia. In addition to anthropogenic factor, the biomass ignition is favored by dangerous
44 consequences of the global climate warming, manifesting in the form of strong temperature, circulation, and
45 hydrologic anomalies (Groisman et al., 2007). In view of enormous stretches of boreal zone, fires occur in
46 some or another region of this vast territory almost every year and influence strongly the radiation budget, air
47 quality, human health, biological diversity, glaciology, etc. To understand the effects of biomass burning on
48 the atmosphere, the physical, chemical and optical properties of smoke particles, it needs to be studied and
49 parameterized with reliable uncertainties.

50 Since 1990s, a large amount of information on characteristics of carbonaceous particles based on in situ
51 measurements and remote sensing using the ground-, aircraft- and satellite-based instruments has been
52 accumulated in hundreds of manuscripts. However, even with the availability of such a volume of data, the
53 determination of key parameters for estimating atmospheric effects of biomass burning is not straightforward,
54 primarily because the smoke properties strongly depend on the set of a variety of reasons, the main of which
55 are the type of biomass, the stage of fire, meteorological conditions at the fire site and on the territory of the
56 dispersal of smoke plumes, age of smoke, etc. (see, e.g., the reviews Dubovik et al., 2002; Reid et al., 2005,
57 2005a; Bond and Bergstrom, 2006; Moosmüller et al., 2009; Giles et al., 2012; Sayer et al., 2014; Nikonovas
58 et al., 2015 and bibliography therein). Another problem is that specific features of individual fires may
59 strongly differ from the ensemble smoke hazes that are a result of some averaging procedures of characteristics
60 of numerous fires. This aspect is crucial for studying the radiation effects of aerosol because, in the framework
61 of regional and global climate models, the most important issue is the development of model representations
62 concerning the aged smoke that dominates regional hazes and affects climate.

63 A stable anticyclone had formed in summer 2012 in Siberian region under the conditions of small-
64 gradient high-pressure baric field (Polyakov et al., 2014), with consequences being that forest and peat fires
65 burned in a few regions of Siberia and encompassed the territory from 1 to 10 million hectares, by different
66 estimates. Optical and microphysical properties of near-ground aerosol and specific features of their vertical
67 structure according to data of in situ measurements, as well as spatiotemporal evolution of aerosol optical
68 depth and active fires according to results of satellite monitoring, are presented in (Gorchakov et al., 2014;
69 Kozlov et al., 2014; Sklyadneva et al., 2015; Vinogradova et al., 2015; Panchenko et al., 2016). In our work,
70 we discuss the columnar optical and microphysical aerosol characteristics, retrieved on the basis of ground-
71 based photometric observations in Tomsk during the period of smoke mist in 2012. The second aim is to
72 compare these results with data of multiyear photometric measurements and satellite observations over the
73 territory of the Western Siberia under the different atmospheric conditions.

74



75 **2 Instrumentation, Sites, and Methods**

76

77 **2.1 Ground-based measurements**

78

79 The measurements, presented in this paper, were made with CIMEL Electronique CE 318 sun-sky radiometer
80 that is a part of AEROSOL ROBOTIC NETWORK (AERONET, (Holben et al., 1998)). These measurements were
81 performed in eastern suburb of Tomsk (“Tomsk”: 56.48°N; 85.05°E) in period of 2003-2010; and since 2011,
82 CE 318 has been operated at “Fonovaya” observatory located 60 km away from the city (“Tomsk-22”:
83 56.42°N; 84.07°E).

84 Direct Sun measurements are made in the spectral channels centered at 340, 380, 440, 500, 675, 870, 940 and
85 1020 nm (bandwidth: 10 nm at full width at half maximum). These solar extinction data are then used to
86 compute aerosol optical depth (AOD, τ_λ) at each wavelength λ except 940 nm, which is used to retrieve total
87 columnar content of water vapor (W). The spectral aerosol optical depth data have been screened for clouds
88 following the methodology of Smirnov et al. (2000). In addition to direct Sun, radiation measurements in solar
89 almucantar are made in four channels of CE 318: 440, 675, 870, and 1020 nm. These data and the inverse
90 automated algorithm of Dubovik and King (2000) (version 1) with enhancements (version 2, Holben et al.,
91 2006) were used to retrieve other aerosol characteristics: volume particle size distribution function
92 $dV/d \ln r (\mu\text{m}^3 \mu\text{m}^{-2})$, scattering phase function, asymmetry factor g_λ , complex refractive index
93 $(n_\lambda - \kappa_\lambda \times i)$, and single scattering albedo ω_λ .

94 Dubovik et al. (2000a) presented uncertainty of retrieval estimates for volume size distribution,
95 refractive index, and single scattering albedo (SSA). For Level 2 data at moderate aerosol loading ($\tau_{440} \sim 0.4$)
96 uncertainties for such retrievals are: 10-35% for the binned size distribution in the intermediate particle size
97 range ($0.1 \leq r \leq 7 \mu\text{m}$); 0.04 and 30-50% for the real and imaginary parts of the refractive index; and 0.03 for
98 single scattering albedo. For typical biomass burning models (Dubovik et al., 2002) these uncertainties were
99 propagated onto the other size distribution parameters (Sayer et al., 2014): the volume mean radius r_v and
100 standard deviation σ of fine (f) and coarse (c) modes are retrieved with errors $0.01 \mu\text{m}$ for r_v^f , $0.2 \mu\text{m}$ for r_v^c ,
101 and 0.06 for σ^f and σ^c . This led to uncertainties of ~ 0.015 – 0.04 in the asymmetry factor (AF) of fine
102 aerosol mode (larger uncertainties at longer wavelengths) and ~ 0.01 in the coarse mode aerosol (smaller
103 uncertainties at longer wavelengths).

104 An original approach, relying upon ground-based spectral measurements of AOD and radiance phase
105 functions, was also used in addition to the algorithm of Dubovik and King (2000) to solve the inverse problem.
106 The first version of the algorithm, namely, Sun-Sky Measurements for Aerosol ReTrieval (SSMART 1.1
107 software package), was implemented under the assumption of (a) the sphericity of aerosol particles and (b) the
108 independence of the complex refractive index on the wavelength and particle size (Bedareva et al., 2013a). An
109 improved version of the algorithm (SSMART 1.2), in which the inverse light scattering problem was solved
110 using the model of a mixture of randomly oriented polydisperse spheroids, was suggested by Bedareva et al.



111 (2014). In both versions, the aerosol optical characteristics are derived in two ways: directly from the spectral
112 Sun-sky radiometer measurements (Way 1) and through the calculation based on the retrieved particle size
113 distribution and complex refractive index (Way 2). On the basis of closed numerical experiments, the accuracy
114 of aerosol retrievals was investigated in error-free conditions and in the presence of measurement errors. The
115 SSMART algorithm was tested as applied to conditions of moderate and increased aerosol turbidity of the
116 atmosphere at Tomsk and Dakar (14 N, 16 W) AERONET sites. It was found that the SSMART and
117 AERONET codes give the consistent estimates of aerosol properties within their retrieval uncertainties
118 (Bedareva et al., 2013ab, 2014).

119 Measured aerosol optical depth and computed retrieval products were used to derive additional aerosol
120 properties. Spectral dependence of AOD is traditionally described by the empirical Ångström formula

$$\tau_{\lambda} = \beta \lambda^{-\alpha}. \quad (1)$$

121 The absorption AOD (AAOD) is calculated for each wavelength using the following equation (Giles et al.,
122 2012)

$$\tau_{abs,\lambda} = \tau_{\lambda} \times [1 - \omega_{\lambda}] \quad (2)$$

123 and, similar to Eq. (1), it can be represented as

$$\tau_{abs,\lambda} = \beta_{abs} \lambda^{-\alpha_{abs}}. \quad (3)$$

124 Analyzing the radiative characteristics of aerosol in the period of 2012 smoke mist, we additionally
125 employed data of multiyear AOD observations at “Tomsk” and “Tomsk-22” sites, obtained from April to
126 October in 2003-2011 and in 2013. Using the method of Kabanov and Sakerin (2006), the total dataset was
127 divided into two subsets: 1) “background” (usual) conditions (998 days); and 2) “ordinary” smoke situations
128 (81 days). By the “ordinary” smokes we mean smoke situations from yearly observed Siberian biomass
129 burning of different types (forest and peat fires, springtime vegetation burning, smokes from remote sources),
130 which were shorter lasting and less severe as compared to 2012 smoke mist. Simultaneous measurements at
131 these sites revealed no statistically significant AOD differences in the warm period of the year (Sakerin et al.,
132 2010); therefore, merging of data, obtained at the neighboring sites (“Tomsk” and “Tomsk-22”), can be
133 regarded as correct.

134 Smaller data volume had gone into comparative analysis of aerosol optical and microphysical
135 characteristics obtained from the inverse procedure. The number of retrievals of total set of characteristics,
136 including refractive index, single scattering albedo, volume size distribution function, and scattering phase
137 function (Level 2, $\tau_{440} > 0.4$), had been 65 in the period of smoke mist (June 15 – August 10, 2012), and 140
138 under the conditions of “ordinary” smokes (April – October 2003-2011, 2013). Out of 140 “ordinary” smoke
139 situations, maximal number of almucantar retrievals had been 39 (2004), 23 (2006), and 22 (2013). In these
140 data, results obtained in May 2004 deserve a special attention. Dry and warm weather, predominating in
141 Novosibirsk and Tomsk regions (with air temperatures exceeding 30°C on separate days) led to numerous
142 forest fires, provoked by burning of last year’s vegetation and bonfires, left unextinguished by fishers and
143 hunters. The number of retrieved aerosol characteristics varied from 1 (2005) to 15 (2008) for remaining
144 period of multiyear observations.



145

146 **2.1 Satellite data**

147

148 AOD observations at 550 nm (collection 6, <http://giovanni.sci.gsfc.nasa.gov/giovanni/>) and Aerosol Small
149 Mode Fraction (collection 5, [http://gdata1.sci.gsfc.nasa.gov/daac-](http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance_id=MODIS_DAILY_L3)
150 [bin/G3/gui.cgi?instance_id=MODIS_DAILY_L3](http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance_id=MODIS_DAILY_L3), valid at time of manuscript preparation) from MODIS
151 instruments were used. These MODIS products (TERRA and AQUA platforms, Level 3 data, i.e., daily
152 averaged within the grid cells $1^\circ \times 1^\circ$) were obtained using the algorithm of Levy et al. (2010).

153

154 **3 General characteristics of the large-scale smoke pollution in the summer of 2012**

155

156 In this section, we present the general characteristics of smoke situation in 2012 on the territory of Tomsk
157 region.

158

159 **3.1 Weather-climate features**

160

161 In summer 2012, a stable anticyclone had formed over the territory of Siberia under the conditions of
162 small-gradient high-pressure baric field (Polyakov et al., 2014), the consequences being substantial changes in
163 climatically significant characteristics and, primarily, an appreciable increase in air temperature and a decrease
164 in precipitation. Data of observations at “Tomsk” meteorological station (WMO_ID=29430, <http://rp5.ru>)
165 indicate that monthly mean temperature in July was maximal over last decade, while precipitation amount was
166 close to a minimum (Figure 1). On the whole, positive anomalies of temperature reached 1.3-7.2°C in June-
167 July on the territory of Tomsk region (~ 56 - 61° N; 75 - 88° E), and precipitation amount was 20-30% of climatic
168 norm. Selyaninov hydrothermal wetting coefficient was also used as a characteristic of wetting (drought)
169 regime (see, e.g., (Polyakov et al., 2014)); it is defined as the ratio of precipitation total for period no shorter
170 than one month to the sum of temperatures for the same period, decreased by a factor of ten. Polyakov et al.
171 (2014) showed that, over the last 70 years, the atmospheric droughts on the territory of Tomsk region were
172 longer than one month in nine summer seasons (the hydrothermal wetting coefficient varied from 0.6 to 1.4),
173 with the summer of 2012 being the driest one.

174

175 These conditions had led to extensive forest fires in the Western and Eastern Siberia (Figure 2),
176 accompanied by considerable pollution of the atmosphere by combustion products (smoke particles, carbon
177 and nitrogen oxides, etc.). Ground-based observations in Tomsk indicate that, in period of maximal smoke
178 pollution on July 25-28, 2012, the aerosol number concentration had been 3000 - 8500 cm^{-3} (particle size 0.4 - 15
179 μm), exceeding the background values by about a factor of 20, while carbon monoxide concentrations reached
180 7.7 ppm (Sklyadneva et al., 2015). Results of satellite (AIRS/TERRA) monitoring indicate that the July-
181 average total CO content over the territory of Tomsk region in 2012 increased by approximately 40% (Fig. 1)
as compared to 2005-2015 (except 2012).



182 Anticyclone over the territory of the Western Siberia persisted for almost two months (second half of
183 June – first decade of August), similar to anomalous situations observed over the European territory of Russia
184 in 1972 and 2010 (Shakina and Ivanova, 2010). At the end of first decade of August, the blocking cyclone
185 started to break, heat rapidly weakened, and air temperature returned to within the climatic norm at all
186 meteorological stations of Tomsk region (Polyakov et al., 2014).

187

188 3.2 Aerosol radiation characteristics

189

190 3.2.1. Ground-based observations

191

192 Figure 3abc shows the time variations in daily average values of AOD, single scattering albedo ω_{440} , and
193 asymmetry factor g_{440} in the summer period of 2012.

194 The AOD data indicate that there are two waves of high atmospheric smoke turbidities: from June 17 to
195 July 5, and from July 19 to August 6. Maximal AOD values (500 nm), reaching 3-5, were observed on July 3-6
196 and July 25-29.

197 The single scattering albedos did not go out of the interval $0.93 \leq \omega_{440} \leq 0.99$ in almost all cases. The
198 asymmetry factor g_{440} varied from 0.66 to 0.74, i.e., in the same range as multiyear average values on the
199 territory of Siberia (Sakerin et al., 2009, 2012, 2014). From results presented here it also follows that, on those
200 days when SSA and AF were retrieved by two methods, the SSMART and AERONET codes gave consistent
201 estimates of aerosol properties within their retrieval uncertainties.

202 Arrival of smoke plumes was also accompanied by a substantial increase in the mass concentrations of
203 black carbon (M_{BC}) and aerosol (M_a) in the near-ground atmospheric layer. Kozlov et al. (2014) showed that
204 the average values of these characteristics at “Fonovaya” observatory in the period from June 17 to August 4
205 varied in the ranges $M_{BC}=1.9-10.3 \mu\text{g m}^{-3}$ and $M_a=103-425 \mu\text{g m}^{-3}$ given the background levels of ~ 0.45 and
206 $\sim 20 \mu\text{g m}^{-3}$ respectively for August 10-31. Similar to AOD, the maximal concentrations of M_{BC} and M_a were
207 observed in the period of July 25-29.

208 The examples of daily average aerosol volume size distribution $dV/d \ln r$ for moderate to high AOD
209 values are presented in Figure 3d. A noticeable salient feature of the $dV/d \ln r$ distribution is the increase in
210 the modal radius and in the width of the mode of finely dispersed fraction during extreme turbidity on July 28
211 as compared to periods on June 27 and July 14, when AOD was much lower.

212 A consequence of anomalously high atmospheric turbidities had been substantial changes in solar
213 radiation reaching the earth’s surface. Figure 4 illustrates the daytime behavior of direct and diffuse radiative
214 fluxes, measured with MS-53 pyrhelimeter and MS-802 pyranometer ($0.305-2.8 \mu\text{m}$) under usual
215 (background) conditions on July 24, 2011 ($\tau_{500} = 0.07$) and in the highly turbid atmosphere on July 28, 2012
216 ($\tau_{500} = 2.14$). Under the influence of smoke plume, there was a two- to four-fold decrease in direct radiation,
217 which was partially compensated for by approximately equal increase in diffuse radiation. The diffuse



218 radiation was considerably (a factor of ~1.8) larger than the direct radiation throughout the day (July 28,
219 2012).

220

221 3.2.2 Satellite observations

222

223 The spatial distributions of AOD at 550 nm and Aerosol Small Mode Fraction ($\eta_{550} = \tau_{550}^f / \tau_{550}$) over
224 Western and partially Eastern Siberia (60°-100°N, 52°-70°E), averaged over the period of June 15 – August 10,
225 2012, are presented in Figure 5. These data show that the territory of the Tomsk region in this period was
226 subjected to intensive influence of large-scale smoke pollution and characterized by high values of AOD and
227 content of fine aerosol fraction: $\tau_{550} > 0.9$; $\eta_{550} > 0.7$.

228

229 4. Discussion of results

230

231 4.1 Temporal and Spectral Variability of Aerosol Optical Depth

232

233 4.1.1. Ground-based data

234

235 Analysis of multiyear AOD variations in a few regions of Russia according to data of photometric
236 observations showed that, after eruption products of Pinatubo volcano had sunk out of the stratosphere, the
237 interannual AOD variations were small and no statistically significant trend was observed in the past two
238 decades (Sakerin et al., 2009, 2012; Sakerin and Kabanov, 2015). For instance, the annually average AOD
239 values (500 nm) in Tomsk during the period of 2003-2015 (except in 2012) varied from 0.13 to 0.21
240 (Figure 6a).

241 Under the influence of massive forest fires in Siberia during summer 2012, the annual AOD value had
242 exceeded the multiyear norm by almost a factor of two and became the highest over the entire period of
243 photometric observations in Tomsk (since 1992) (Sakerin and Kabanov, 2015). Average summertime AOD
244 values changed even stronger: in particular, the average value τ_{500} in July had increased by a factor of 4.5 as
245 compared to multiyear data (Figure 6b). Even if we omit anomalously high turbidities ($\tau_{500} > 1$), which may
246 be partly due to clouds invisible through haze, the average AOD values in summer months of 2012 go out of
247 the corridor defined by standard deviations (SDs).

248 More detailed characteristics of atmospheric AOD for three wavelengths in UV, visible, near IR ranges
249 during the smoke mist in comparison with the multiyear average data for July are presented in Table 1. Also
250 given are the average characteristics for various situations of “ordinary” smokes, which are observed every
251 year in Siberia in warm period. The comparison showed (see also Figure 6c) that AOD during smoke mist
252 exceeds the background values by a factor of 5.5-6 in the entire spectral range. In absolute value, AOD
253 increased stronger in shortwave part of spectrum, i.e., due to finely dispersed aerosol. The slight increase in
254 Ångström exponent α , which depends on the interrelation between contributions of fine and coarse aerosols to



255 AOD, also indicates that small particles predominate in smoke aerosol. Despite the temperature, aerosol, and
256 other anomalies, the total water vapor content of the atmosphere in the period of smoke mist differed little
257 from the multiyear norm.

258 Average AOD characteristics in “ordinary” smokes incorporate data for fires of different types and
259 distances from the region of measurements and, as such, occupy an intermediate position between background
260 conditions and smoke mist. Independent of their intensity (mist or “ordinary” smokes), a common feature of
261 all smokes is an identical exponent α , which characterizes higher content of fine aerosol.

262 It is noteworthy that the multiyear average values of α and AOD in the near-IR range, calculated for
263 background conditions and total dataset, differ little (see columns 2 and 3 in Table 1), primarily because of
264 relatively small number (about 8%) of smoke situations in the total dataset.

265 Under the assumption that aerosol size distributions are bimodal, O’Neil et al. (2001, 2003) have
266 developed a spectral deconvolution algorithm (SDA) to infer the component fine and coarse mode optical
267 depth from spectral dependence of AOD. In June–August 2012, the coarse mode τ_{500}^c is less than fine mode
268 τ_{500}^f and is typically low (only 3 days show that $\tau_{500}^c \sim 0.2$), whereas τ_{500}^f is high and exhibits very large day-
269 to-day variability (Figure 7a). The parameter η_{500} , which characterizes the contribution of fine fraction to
270 AOD at 500 nm, exceeded 0.7 and approached 0.9 at $\tau_{500} \geq 0.6$ (Figure 7b). These results are consistent with
271 earlier published data, according to which content of finely dispersed aerosol predominately increases during
272 vegetation burning in the absence of any other significant sources (Reid et al., 2005a; Eck et al., 2009; Giles et
273 al., 2012).

274

275 4.1.2 Analysis of satellite observations

276

277 Analysis of spatiotemporal AOD variations was confined to the consideration of the central part of the
278 Western Siberia. A salient feature of this territory is a uniform landscape (without mountainous terrain) and
279 the absence of large sources of anthropogenic pollution. Under usual conditions, the AOD values, retrieved
280 from satellite observations, are characterized by small (background) values and by quite a uniform spatial
281 distribution (Zhuravleva et al., 2009a; Sakerin et al., 2012).

282 Since the formation and evolution of smoke plumes are strongly affected by large-scale atmospheric
283 dynamics, spatial distributions of AOD may substantially change from one day to another. The main specific
284 features of variations can be identified through analysis of AOD fields, averaged for a few 5-day periods,
285 selected by taking into account the ground-based observations (Fig. 3a): on June 6–10 there were background
286 conditions (before beginning of forest fires); on July 1–5 and July 24–28 there were maximal smoke turbidities;
287 and on July 10–14 there were relatively low AOD values observed between two turbidity maxima. Hereinafter,
288 for calculations of the AOD values averaged over the selected time interval (5 days) we used the daily
289 satellite product.

290 Spatial AOD distributions, presented in Figure 8, show that the “Tomsk-22” observation site was either
291 at the center (Fig. 8b) or on the periphery of smoke plumes (Fig. 8a) in different periods of time. The periods



292 of small turbidities of the atmosphere (on June 6-10 and July 10-14) were characterized by quite a uniform
293 AOD distribution, and the largest spatial inhomogeneities and values $\tau_{550} > 1$ were observed over the central
294 part of the Western Siberia on July 24-28 (Fig. 8 and Table 2).

295 The statistical characteristics in Table 2 were calculated using measurements from AQUA and TERRA
296 platforms (MODIS collection 6, <http://giovanni.sci.gsfc.nasa.gov/giovanni>). These data suggest that, for three
297 out of four periods considered here, the average AOD values at “Tomsk-22” site are close to spatiotemporal
298 averages on the entire territory of the Western Siberia: the difference between satellite and ground-based data
299 is much less than standard deviations, except in period of July 24-28, when the average AOD at “Tomsk-22”
300 site had been $\tau_{550} = 2.74$ and exceeded the average value for the Western Siberia (MODIS) $\tau_{550} = 1.06$. At
301 the same time, the satellite data in the ($1^\circ \times 1^\circ$) region of “Tomsk-22” site well agree with results of photometric
302 observations. Thus, the results of ground-based measurements in the region of “Tomsk-22” can be considered
303 to reflect not only the local AOD features, but also the regularities of variations for the entire territory of the
304 Western Siberia.

305

306 4.2. Retrieval results

307

308 We will compare the retrievals of aerosol optical and microphysical characteristics under the conditions of
309 smoke mist in 2012 against the average data for ordinary smokes (Table 3). Since the AOD dependence of C_v
310 and r_v for fine and coarse aerosol modes has previously been noted for a wide variety of aerosol types
311 (including biomass burning aerosol (Dubovik et al., 2002; Sayer et al., 2014)), Table 3 also presents the
312 resulting linear regression relationships between the pairs $(C_v^{f(c)}, \tau_{440})$ and $(r_v^{f(c)}, \tau_{440})$, as well as the
313 corresponding linear correlation coefficients R .

314

315 4.2.1 Volume size distributions

316

317 During the period of smoke mist in 2012, the average values of volume median radii for finely and coarsely
318 dispersed fractions were $r_v^f = 0.18 \mu\text{m}$ and $r_v^c = 3.3 \mu\text{m}$, a little larger than these characteristics for ordinary
319 smokes, equaling $0.16 \mu\text{m}$ and $2.9 \mu\text{m}$ respectively (Table 3). The r_v^f value, in both cases, was in the range of
320 values, obtained for biomass burning in other regions of the globe (Reid et al., 2005, 2005a; Eck et al., 2009;
321 Chubarova et al., 2012), as well as in the range of measurements for aged smoke, performed using Optical
322 Particle Counter ($0.1 - 3 \mu\text{m}$) and Differential Mobility Particle Sizer ($0.01 - 0.6 \mu\text{m}$) in boreal zones of
323 Europe and North America (Reid et al., 2005). Like in Siberia, the wider size distribution of fine particles than
324 for ordinary smokes (Table 3; Fig. 3d) was also noted by other authors for severe fires in boreal zone of
325 Alaska (Bonanza Creek, 2004-2005, Eck et al., 2009).

326 The noteworthy salient features of finely dispersed fraction during summer 2012 are as follows. The
327 contribution of finely dispersed fraction to the total volume (C_v^f/C_v^t), on the average, increased to



328 0.8 ± 0.065 as compared to value 0.65 ± 0.21 according to multiyear observations. Volume concentrations and
329 volume median radii of the fine mode are plotted in Figure 9a as functions of AOD at 440 nm. This figure
330 shows that there is general increase in volume concentrations C_v^f as τ_{440} increases (Fig. 9a). The median
331 radius of the fine mode increased from about 0.15 to 0.22 μm as τ_{440} changed from 0.4 to 1.5 and larger (Fig.
332 9b). The smoke particles have large radii, and r_v^f and τ_{440} are closely correlated, possibly because of high
333 concentration of aerosol, which leads to greater coagulation, condensation, and gas-to-particle conversion
334 (Reid et al., 1998, 2005; Eck et al., 2009). From this figure it also follows that the interrelation between
335 median radius of finely dispersed fraction and AOD at 440 nm in ordinary smokes is much less pronounced,
336 primarily because of the variety of properties biomass burning aerosol combined in this dataset. For instance,
337 in May 2004, the median radius was relatively small ($r_v^f \sim 0.14 \mu\text{m}$) and did not practically change with
338 varying τ_{440} . Most probably, this was because burning of last year's vegetation was one of the sources of
339 smoke particles in this period of time. (An analogous feature was also observed during African savanna fires
340 (Dubovik et al., 2002)).

341 In contrast to data of multiyear observations, in summer 2012 an increase in the volume concentration
342 and median radius of the coarse mode was observed with the growing AOD. Most probably, soil particles,
343 being suspended by saltation of surface dust driven by fire generated winds, were also present during this
344 period of time in the composition of coarse mode in addition to carbon aggregates.

345 Results of retrieval of disperse composition of finely dispersed aerosol in the atmospheric column agree
346 with measurements in near-ground layer (Kozlov et al., 2014). Data of spectral-polarization nephelometric
347 measurements were used to show that the volume median radius of the fine mode under the conditions of weak
348 turbidity of the atmosphere (July 10–13, 2012) had increased from 0.1 μm to 0.4–0.5 μm when smoke plumes
349 intruded to the region of observations (July 25–29, 2012).

350

351 4.2.2 Refractive index

352

353 Average values of the real and imaginary parts of refractive index under the conditions of smoke mist and
354 ordinary smokes are presented in Table 3. In ordinary smokes, the imaginary part of refractive index κ_λ
355 changes weakly with the increasing wavelength and is approximately 0.01. However, the imaginary refractive
356 index in the Tomsk smoke mist shows low values and relatively large decrease in κ_λ as the wavelength grows
357 from 440 to 675 nm. The κ_λ variations in the wavelength interval of 675–1020 nm are not as strong.

358 The quantitative differences and specific features of spectral dependence of the imaginary refractive
359 indices stem from different properties of atmospheric carbonaceous particles, i.e., black carbon (BC) and
360 organic aerosol (OA). Recent studies showed that OA components can contribute substantially to light
361 absorption. In contrast to BC, which absorbs light throughout the UV-visible spectrum, such an OA
362 component as “brown carbon” (BrC) absorbs mostly in the ultraviolet wavelength and less significantly in the
363 visible spectral range (Kirchstetter et al., 2004; Bergstrom et al., 2007; Chen and Bond, 2010; Zhong and



364 Jang, 2014). The majority of BrC is emitted to the atmosphere through low temperature, incomplete
365 combustion of biomass, bio- and fossil fuel (Bond, 2001; Kirchstetter et al., 2004; Bergstrom et al., 2007;
366 Lewis et al., 2008; Chen and Bond, 2010; Zhong and Jang, 2014).

367 The absorption efficiency and spectral dependence vary depending on the type of BrC origin. Lu et al.
368 (2015) reviewed available measurements (laboratory and field observations) of light-absorbing primary
369 organic aerosols, and quantify the wavelength-dependent imaginary refractive indices (κ_{OA}) for the bulk
370 primary OA emitted from biomass/biofuel, lignite, propane and oil combustion sources. Based on generalized
371 information, they suggested to parameterize κ_{OA} of biomass/biofuel combustion sources as a function of BC-
372 to-OA ratio. Analysis of imaginary refractive indices showed the stronger wavelength-dependent κ_{OA} for
373 lower BC-to-OA ratio conditions (smoldering combustion), while the greater κ_{OA} values at $\lambda > 350$ nm are
374 observed for higher BC-to-OA ratio (flaming combustion).

375 These results allow us to hypothesize that the reason for the κ_{λ} decrease in the interval of 440-675 nm
376 during summer 2012 may be due to the contribution of compounds absorbing radiation in UV wavelength
377 region (“brown” carbon). To confirm this hypothesis, we considered OMI observations of the Ultraviolet
378 Aerosol Index (UVAI), the value of which is sensitive to aerosol absorption in the ultraviolet wavelength
379 region (Torres et al., 1998; Jethva and Torres, 2011; Hammer et al., 2016)]. Satellite data indicate that UVAI
380 values were quite high in the period of smoke mist, and exceeded 4-5 on individual days, thus explaining the
381 above-mentioned κ_{λ} decrease in going from UV to visible spectral region (Figure 10).

382 The relatively constant imaginary refractive index in ordinary smokes owes to averaging over many
383 situations, differing, in particular, in the type of burning biomass (peat, forest, grass). For instance, in May
384 2004, when the characteristics of smoke aerosol were quite homogeneous, the average spectral behavior of κ_{λ}
385 was weakly manifested, with $\kappa_{\lambda} \approx 0.013$ in the interval of 440-1020 nm. At the same time, the UVAI value
386 did not exceed 1.2 (<http://giovanni.sci.gsfc.nasa.gov/giovanni/>), suggesting that BC was seemingly the main
387 absorbing substance in this period of time.

388

389 4.2.3 Single scattering albedo

390

391 Detailed analysis of spectral SSA in 2012 has revealed two types of dependence: monotonically decreasing
392 and monotonically increasing ω_{λ} with the growing wavelength (Figure 11).

393 The main group of SSA data (45 cases out of 65) is characterized by a weakly decreasing spectral
394 dependence: the range of the differences $\Delta\omega_{\lambda} = \omega_{440} - \omega_{1020}$ did not exceed 0.03 (Fig. 11a). Practically all set
395 of retrieved SSA of this type lies between the curves 2 ($\omega_{440} = 0.926$) and 3 ($\omega_{440} = 0.996$). In other cases,
396 the ω_{λ} decrease with the growing wavelength is larger, but does not exceed 0.06 (curve 4, Fig. 11a). We note
397 that the values of the imaginary part of refractive index are ~ 0.006 for this subset of data.



398 In a few (14 out of 65) situations with high atmospheric turbidities ($\tau_{440} > 0.9$), there was an increase
399 in SSA with the growing wavelength (curve 1 in Fig. 11a). The monotonic increase in ω_λ agrees with
400 decrease in κ_λ (on the average, from 0.01 at 440 nm to ~ 0.006 at 675 nm), presumably due to additional
401 absorption of “brown” carbon in the UV wavelength region.

402 On the whole, the 2012 smoke mist was characterized by weak spectral variations in SSA, the average
403 being ~ 0.96 , consistent with previous results in other regions of Eurasian boreal zone: in Moscow (2002 and
404 2010) and Alaska (2004–2005), where the smoldering phase prevailed over the flaming fire phase (Eck et al.,
405 2009; Chubarova et al., 2011; Sayer et al., 2014). The relatively constant spectral single scattering albedo of
406 smoke mist in 2012 may be also partly due to growth of aerosol particle sizes (Subsect. 4.2.1, Table 3; see also
407 Eck et al., 2009).

408 The SSA retrievals in atmospheric column satisfactorily agree with data in the ground-layer (Kozlov et
409 al., 2014), according to which the single scattering albedos in the visible range ($\lambda=510$ nm) were in the interval
410 of 0.95–0.98. At the same time, the imaginary and real parts of the complex refractive index of dry base of
411 substance varied in the ranges of 0.01–0.02 and 1.36–1.47 respectively.

412 Overwhelming majority of results of individual retrievals in “ordinary” smokes also showed a
413 monotonic SSA decrease with the increasing wavelength, with the opposite dependence being observed in
414 only 6 cases out of 140. However, in contrast to smoke mist, the spectral SSA behavior was stronger
415 pronounced: $\Delta\omega_\lambda$ reached 0.06–0.18 in 18% of cases; $\Delta\omega_\lambda$ was 0.03–0.06 in 40% of cases, and $\Delta\omega_\lambda$ was
416 less than 0.03 in 34% of cases.

417 The average single scattering albedos in “ordinary” smokes, presented in Fig. 11b, turned out to be
418 about 0.02 lower than those observed in boreal forests of USA and Canada (Dubovik et al., 2002). These
419 differences may be both due to methodic and physical factors. Methodic-type differences might stem from
420 different methods for selecting the smoke situations. The specific features of the spectral dependence of SSA
421 can also be explained by different combinations of the composition and age of smoke aerosol, which were
422 recorded at AERONET observation sites. This is clearly illustrated by results obtained in May 2004 in Toms
423 (Fig. 11b). Lower values and pronounced spectral variations of SSA (a decrease from 0.9 to 0.85) may be due
424 to the last year’s grass and agricultural vegetation combustion products, present in the smoke composition, as
425 well as due to flaming phase of combustion which predominated in that period of time. (Close values and
426 analogous spectral dependence of single scattering albedo were also noted in African savanna fires (Zambia),
427 Dubovik et al., 2002, Sayer et al., 2014).

428 Aerosol absorption optical depth $\tau_{abs,\lambda}$ and absorption Ångström exponent α_{abs} , computed for the
429 spectral interval of 440–870 nm (Sect. 2), were considered as other characteristics of absorbing properties of
430 aerosol particles.

431 Data, presented in Fig. 11d, show that the average values of $\tau_{abs,\lambda}$ in the period of smoke mist were
432 about a factor of 1.5–2 smaller than for “ordinary” smokes. In addition, there are differences in α_{abs} , which
433 are 1.6 ± 0.44 (smoke mist) and 1.2 ± 0.5 (ordinary smoke) respectively.



434 The AAE values can be used as an indicator of aerosol composition (Andreae and Gelencsér, 2006;
 435 Russell et al., 2010; Schuster et al., 2016). The relatively low values $\alpha_{abs} \sim 1$ are typical for aerosol absorption
 436 largely dominated by black or light absorbing carbon, while larger values suggest absorption by different
 437 material organic carbon (Bergstrom et al., 2002, 2007; Kirchstetter et al., 2004; Lewis et al., 2008; Russell et
 438 al., 2010; Schuster et al., 2016). The α_{abs} increase in the 2012 fires may indicate that, on the average, smoke
 439 absorption possibly shifted from domination by black carbon in “ordinary” smokes to an increasing influence
 440 of absorption by other materials most likely organic carbon (see also Fig. 10).

441 In contrast to single scattering albedo, the spectral asymmetry factor differs insignificantly among the
 442 2012 smoke mist, “ordinary” smokes, and May 2004 smokes (Table 3, Figure 11c).

443

444 5. Radiative effects of smoke aerosol

445

446 The radiative effects of smoke aerosol were accounted for by calculating the transmittance (T), albedo (A), and
 447 absorptance of the atmosphere (ABS_{ATM}) and underlying surface (ABS_{SUR}):

$$T = 100\% \times F_{BOA}^{\downarrow} / F_{TOA}^{\downarrow}, \quad A = 100\% \times F_{TOA}^{\uparrow} / F_{TOA}^{\downarrow}, \quad (4)$$

$$ABS_{ATM} = 100\% \times (F_{TOA}^{\text{net}} - F_{BOA}^{\text{net}}) / F_{TOA}^{\downarrow},$$

$$ABS_{SUR} = 100\% \times F_{BOA}^{\text{net}} / F_{TOA}^{\downarrow}.$$

448 Here, $F_{BOA(TOA)}^{\downarrow(\uparrow)}$ are broadband solar radiative fluxes in the interval of (0.2-5 μm), the symbols $\downarrow(\uparrow)$ are used
 449 to denote the downward and upward radiation at the top of the atmosphere (TOA) and bottom of the
 450 atmosphere (BOA). Radiative influxes F^{net} at different levels are calculated as follows

$$F_{BOA(TOA)}^{\text{net}} = F_{BOA(TOA)}^{\downarrow} - F_{BOA(TOA)}^{\uparrow} \quad (5)$$

451 The results of radiative flux simulation were used to calculate the direct radiative effect (DRE) at TOA
 452 and BOA and in atmospheric column:

$$\Phi_{TOA(BOA)} = F_{TOA(BOA)}^{\text{net},a} - F_{TOA(BOA)}^{\text{net},R}, \quad \Phi_{ATM} = \Phi_{TOA} - \Phi_{BOA}, \quad (6)$$

453 where the superscripts “a” and “R” correspond to the calculations in aerosol-molecular atmosphere and in the
 454 aerosol-free atmosphere, with only molecular (Rayleigh) scattering and absorption taken into consideration.
 455 The negative and positive DRE values are associated with an aerosol cooling and warming, both at TOA and
 456 BOA. In addition to the proper DRE, we also considered the radiative effect efficiency

$$\Phi_{TOA(BOA)}^e = \Phi_{TOA(BOA)} / \tau_{550}, \quad \Phi_{ATM}^e = \Phi_{ATM} / \tau_{550}, \quad (7)$$

457 which characterizes the rate at which the atmosphere is forced per unit of AOD at 550 nm at TOA and BOA.

458 The Φ^e value depends on reflective properties of underlying surface, size distribution of aerosol particles, and
 459 their chemical composition; also, it depends on AOD, to some extent (due to the multiple scattering effects).

460

461 5.1 Model and input data



462

463 The broadband fluxes of the solar radiation in the molecular-aerosol plane-parallel atmosphere were calculated
 464 using the algorithm of the Monte Carlo method, which we developed earlier (Zhuravleva et al., 2009a). The
 465 radiative fluxes at a given atmospheric level z are represented as a sum of fluxes in separate wavelength
 466 intervals:

$$F^{\downarrow(\uparrow)}(z) = \sum_{i=1}^M F_i^{\downarrow(\uparrow)}(z), \quad (8)$$

467 where $M=31$ is the number of bands $\Delta\lambda = (\lambda_i, \lambda_{i+1})$, $i = 1, \dots, M-1$, $\lambda_1 = 0.2 \mu\text{m}$, $\lambda_M = 5.0 \mu\text{m}$. Within each
 468 subinterval, the optical characteristics of aerosol and molecular scattering coefficient are assumed to be
 469 constant and equal to their values in the middle of subinterval. The transmission function is approximated by
 470 a finite exponential series (k-distribution method). The algorithm intrinsically takes into account the multiple
 471 scattering, absorption by aerosol and molecular particles, as well as reflection of incident radiation from the
 472 underlying surface according to Lambert law. The comparisons showed that the numerical simulation results
 473 are in a satisfactory agreement with results of line-by-line calculations and data of field measurements
 474 (Tvorogov et al., 2008; Zhuravleva et al., 2009a, 2014).

475 The spectral AOD (340-1020 nm), the water vapor content, the column-integrated single scattering
 476 albedo, and the asymmetry parameter taken from AERONET were used as the main input parameters of the
 477 algorithm under the smoke conditions. In the interval of 440-1020 nm, the spectral values ω_λ and g_λ have
 478 been linearly interpolated from the values of SSA and AF retrieved at the four AERONET inversion
 479 wavelengths, while for $\lambda \leq 440$ nm and $\lambda \geq 1020$ nm they have been considered constant, similar to Garsia
 480 et al.(2012) and Panchenko et al. (2012). Under the background conditions, Level-2.0 retrieval products for
 481 the single scattering albedo and asymmetry factor, obtained on the basis of standard AERONET algorithm, are
 482 not available due to low AOD values ($\tau_{550} = 0.13$ according to data of multiyear ground-based measurements
 483 under summer conditions, Sakerin et al., 2009; Sakerin and Kabanov, 2015). Therefore, OPAC model
 484 (averaged continental aerosol, relative air humidity is 70%, (Hess et al., 1998) was used to simulate the
 485 radiative characteristics under the conditions of the weakly turbid atmosphere.

486 As in (Panchenko et al., 2012), the aerosol optical depth was assumed to be constant outside the
 487 wavelength interval of 340-1020 nm: $\tau(\lambda \leq 340 \text{ nm}) = \tau_{340}$, $\tau(\lambda \geq 1020 \text{ nm}) = \tau_{1020}$. Such an approach was
 488 chosen for the following reasons. The contribution of solar radiation, incoming at TOA in the interval of 200-
 489 340 nm, is about 3.5%; therefore, in the absence of measurements, the specified character of spectral
 490 dependence of AOD cannot influence significantly the simulation results. Based on the data of multiyear
 491 ground-based observations on the territory of Siberia, Sakerin and Kabanov (2007) showed that AOD hardly
 492 varies in the interval $\lambda > 1000$ nm; therefore, we can assume that $\tau(\lambda > 1000 \text{ nm}) \sim \tau(\lambda \sim 1000 \text{ nm})$.

493 The molecular absorption coefficients were calculated on the basis of HITRAN2008 database and
 494 MT_CKD v.2.4 continuum model (http://rtweb.aer.com/continuum_frame.html), using a regional model of
 495 temperature, pressure, and water vapor concentration profiles (Komarov and Lomakina, 2008), taking into
 496 account the absorption by all atmospheric gases, which was presented in the AFGL meteorological model



497 (Anderson et al., 1986). The profiles of ozone O_3 and carbon dioxide CO_2 were specified taking into
498 consideration the data of multiyear observations, obtained on the territory of the Western Siberia during
499 summer period: the total O_3 content was taken to be equal to 340 DU (according to data of TOMS satellite
500 instrumentation, 2000-2010), and the CO_2 mixing ratio was taken to be equal to 380 ppm (according to data of
501 aircraft sensing in 1997-2007 (Arshinov et al., 2009)).

502 It is well known that the surface albedo affects (primarily upward) radiative fluxes and may even cause
503 DRE sign reversal (see, e.g., Zhuravleva et al., 2009b; Garsia et al., 2012; Tomasi et al., 2015). The processes
504 of soot sedimentation on the ground and appearance of blanked patches from burning produce changes in the
505 surface reflectance and possible variations in the radiative characteristics of the atmosphere. A detailed
506 discussion of these problems is beyond the scope of the present work; therefore, all calculations below were
507 performed for the same surface albedos $A_{s,\lambda}$, specified using multiyear data of MODIS satellite
508 measurements for the region of Tomsk in June – August (Moody et al., 2005).

509 Data of Fontenla et al. (1999) were used to account for the spectral behavior of the solar constant.

510 The applicability of the algorithm and the approach to specifying the set of input parameters was
511 confirmed by our earlier results of the complex radiation experiments (Zhuravleva et al., 2009a, 2014).

512

513 5.2 Simulation results

514

515 Recent studies showed that consideration of only specific spectral range of aerosol properties and neglect of
516 uncertainties in specifying the input parameters (aerosol optical characteristics, surface albedo, total content
517 and vertical profiles of concentrations of atmospheric gases, etc.) may be important error sources in estimates
518 of aerosol radiation effects (Myhre et al., 2003; Zhou et al., 2005; Garcia et al., 2008, 2012; Zhuravleva et al.,
519 2009ab). Magnitude of these errors depends mainly on aerosol type (background continental, oceanic, biomass
520 burning, urban-industrial, desert dust, etc.), reflection model, and albedo of underlying surface, as well as on
521 solar zenith angle (see, e.g., Garsia et al., 2008, 2012, 2014; Zhuravleva and Sakerin, 2009b).

522 In this work, we present the *diurnally* average radiative effects for 5 different situations (Table 4). To
523 estimate correctly their uncertainties, it is necessary to take into account both variations in the characteristics
524 of the atmosphere and underlying surface during the day and the dependence of uncertainties on the solar
525 zenith angle. The estimates, presented in the literature, were generally obtained for fixed illumination
526 conditions (see, e.g., Garsia et al., 2008, 2012, 2014; Zhuravleva and Sakerin, 2009b), while uncertainty
527 estimates for *diurnally* average radiative effects are scarce (see, e.g., Tomasi et al., 2015, Esteve et al., 2016).
528 In this work, we restrict ourselves to a discussion of just radiation effects in different situations. Issue of what
529 is the magnitude of uncertainty due to inaccurate information on input parameters of the problem needs
530 additional numerical experiments and is beyond the scope of the present work.

531 Average values of AOD, W, SSA, and AF, which reflect the “average” radiative effects of smoke and
532 background aerosol, were chosen as input parameters for the first three cases. In order to avoid the effect of
533 astronomical factor, the instantaneous values of $F_{TOA(BOA)}^{\downarrow(\uparrow)}$ were calculated for period between sunrise and



534 sunset on July 15 for the latitude of Tomsk (56° N). Cases 4 and 5 correspond to the conditions of strong (July,
535 27) and relatively weak (July, 14) aerosol turbidity and, as such, make it possible to estimate the variability
536 range of radiative characteristics in the period of smoke mist.

537 Figure 12 shows how radiation characteristics of the atmosphere and underlying surface are
538 redistributed under different conditions.

539 The atmospheric transmittance (and, hence, ABS_{SUR}) was maximal (71%) under the background
540 conditions. The appearance of optically dense smoke cloud had a consequence that T decreased to 60% (Case
541 3). Immediately in the period of 2012 smoke mist, the T value varied from 67% (July 14) to 37% (July 27),
542 being almost a factor of two smaller in the latter case than under the background conditions. Data of
543 calculations agree with results of measurements of total radiative fluxes in Tomsk (Sklyadneva et al., 2015),
544 which indicated that on July 27, 2012 the total radiation decreased by about 50% relative to the usual
545 conditions. Atmospheric albedo was also maximal (34%) on this day.

546 In analysis of ABS_{ATM} , the existence of two competing factors should be taken into consideration. If
547 we represent the atmospheric absorption as a series in the order of scattering, its n th term will be proportional
548 to $\omega_\lambda^{n-1}(1-\omega_\lambda)$; and for large ω_λ the contribution of high orders of scattering will be significant. Therefore,
549 AOD increase (and, consequently, increment in the average order of scattering) favors absorption growth due
550 to the contribution of high orders of scattering. At the same time, large SSA values act as a factor reducing the
551 atmospheric absorptance.

552 The ABS_{ATM} value, calculated with averaged parameters, increased from 22% (Case 1) to 26%
553 (Case 3); while for certain situations during summer 2012 the atmospheric absorptance varied in the range of
554 24-36%. A substantial increment in ABS_{ATM} on July 27 had been a consequence of a considerable (almost an
555 order of magnitude) increase in AOD, accompanied by growth of the average order of scattering.

556 The direct radiative effect of aerosol for fixed characteristics of underlying surface and fixed solar
557 zenith angle primarily depends on AOD, single scattering albedo, and asymmetry factor (Zhou et al., 2005;
558 Yu et al., 2006; Tomasi et al., 2015). The results, presented in Fig. 12b, show a cooling effect of aerosol at the
559 top and bottom of the atmosphere. As expected, the interrelation between DRE values was determined by
560 aerosol optical depth and had been maximal (in absolute value) on July 27, 2012: $\Phi_{BOA} = -150 \text{ W m}^{-2}$,
561 $\Phi_{TOA} = -75 \text{ W m}^{-2}$, $\Phi_{ATM} = 75 \text{ W m}^{-2}$. Under the background conditions, the DRE value was minimal: -
562 13 W m^{-2} at BOA and -5 W m^{-2} at TOA.

563 The AOD influence on direct radiative effect efficiency Φ^e , as compared to DRE, is much less
564 pronounced, making it possible to estimate the influence of absorbing and scattering aerosol properties on
565 radiative effects. However, it should be kept in mind that the increase of multiple scattering effects and
566 attenuation of the transmitted radiation for large AOD moderate their effect (Conant et al., 2003).

567 For small atmospheric turbidity, the aerosol single scattering albedo weakly influences the radiative
568 effect efficiency, and high $|\Phi^e|$ values at TOA, BOA, and in the atmosphere are primarily determined by AOD



569 (Case 1, Fig. 12c). The increase of efficiency for lowest AOD range was also noted by other authors (see, e.g.,
 570 Garcia et al., 2012; background continental regions).

571 The aerosol optical depths in the ordinary smokes and on July 14 are close in value, whereas SSA
 572 values substantially differ (Table 4). A consequence of this is the inequality $|\Phi^e(\text{Case 2})| > |\Phi^e(\text{Case 4})|$,
 573 primarily because of higher absorptance of aerosol particles in ordinary smokes. Comparison of cases 3-5
 574 shows that the $|\Phi^e(\text{Case 4})|$ value is maximal for SSA close to unity (Fig. 12c), suggesting that the influence
 575 of absorbing and scattering aerosol properties on radiative effect efficiency is moderated for high AOD values.

576

577 6 Conclusion

578

579 Previous studies showed that smokes from vegetation burning, together with large volcanic eruptions, are most
 580 intense natural sources of aerosol-gas emissions in boreal zones of the planet; they strongly influence the
 581 radiation budget on the scales of large regions a few weeks long. One such event, i.e., smoke mist due to
 582 massive forest fires, had taken place during summer 2012 in a few Siberian regions.

583 In this work, we present the results of complex study of the optical and microphysical characteristics
 584 and radiation effects of aerosol, observed under the conditions of severe smoke turbidity of the atmosphere.
 585 The basis for analysis had been photometric observations (AERONET/Tomsk-22) of spectral solar radiation
 586 with the use of the algorithms of solution of inverse problems of atmospheric optics and model calculations of
 587 the main components of shortwave radiation budget. The obtained radiation characteristics in smoke mist
 588 situation are compared with data of multiyear (2003-2013) observations under the background conditions, for
 589 “ordinary” smokes, as well as with results of studying the smoke aerosol obtained by other authors.

590 Intensive wildfires in 2012 in Siberia had led to extremely high aerosol loading of the atmosphere: the
 591 average AOD(500 nm) in the period of smoke mist had been 0.95 ± 0.86 , a factor of 6 larger than under the
 592 background conditions (0.16 ± 0.08), and almost a factor of 2.5 larger than AOD in “ordinary” smokes
 593 (0.36 ± 0.18). The AOD value exceeded 3 in certain periods of measurements (on July 24-28).

594 Like at other AERONET sites, where smoke aerosol was recorded, in Tomsk the volume aerosol size
 595 distributions were bimodal with dominating fine-mode fraction. In June-August 2012, the mean median radius
 596 of fine fraction r_v^f had increased to $0.18 \mu\text{m}$ as compared to “ordinary” smokes ($r_v^f = 0.16 \mu\text{m}$). The width
 597 of the fine mode distribution increased, as was the case in period of intense fires during summer 2004-2005 in
 598 Alaska [Eck et al., 2009]. In contrast to data of multiyear observations, in summer 2012 an increase in the
 599 volume concentration and median radius of the coarse mode was observed with the growing AOD.

600 The average imaginary refractive index of the Tomsk smoke mist shows low values and relatively large
 601 decrease in κ_λ as the wavelength grows from 440 nm (0.0067) to 675 nm (0.0054). The κ_λ variations in the
 602 wavelength interval of 675-1020 nm are not as strong. At the same time, in ordinary smokes the imaginary part
 603 of refractive index showed spectral behavior close to neutral one ($\kappa_\lambda \approx 0.01$).

604 A consequence of small values of the imaginary index of refraction coupled with the large fine-mode



605 particle radius had been, on the average, quite high single scattering albedos of smoke aerosol, weakly varying
606 with wavelength (0.96). A similar situation was also observed at other AERONET sites, located in boreal zone
607 of Eurasia, during extended wildfires: Moscow (2002 and 2010), Alaska (2004 and 2005). At the same time,
608 increasing spectral behavior of single scattering albedo with the growing wavelength was observed in certain
609 periods of smoke turbidities. Possibly, this spectral dependence was because “brown” carbon, which absorbs
610 most intensely in ultraviolet spectral region, was present in the atmosphere. A comparative analysis also
611 showed that SSA values and their spectral dependence differ between smoke mist and “ordinary” smokes. In
612 the latter case, the increase in absorptance of aerosol particles and decrease in SSA with the growing
613 wavelength well correspond to the character of SSA variations, recorded in boreal zone of USA and Canada
614 according to data of multiyear observations (Dubovik et al., 2002).

615 Such extraordinary events as severe smoke mist (Siberia, 2012; Moscow, 2010; etc.) are quite rare in
616 occurrence. However, when optical and microphysical characteristics, obtained in these periods of time, are
617 included in the total dataset (ordinary smokes, usual conditions), this may introduce substantial changes in the
618 statistical characteristics of aerosol, typical for a given region (see, e.g., Sayer et al., 2014), and will lead to a
619 greater uncertainty in estimates of radiation-climatic effects of aerosol. In our opinion, it is more correct to use
620 the obtained information for analysis of characteristics of “pure” smoke aerosol (in view of its predominating
621 contribution), as well as for estimation of maximal radiation effect of smokes.

622 The results of simulating the diurnally average radiative characteristics, presented in the work, reflect
623 the “average” radiative effects of smoke and background aerosol. As compared to background conditions and
624 “ordinary” smokes, under the conditions of smoke mist the cooling effect of aerosol intensifies (predominately
625 due to a substantial increase in AOD): direct radiative effects at the bottom and top of the atmosphere are -13,
626 -35, and -60 W m⁻², and -5, -14, and -35 W m⁻² respectively. Values of direct radiative effect efficiency Φ^e
627 under the background conditions and under the conditions of ordinary smokes are comparable ($\Phi_{BOA}^e \sim 100$
628 W m⁻², $\Phi_{TOA}^e \sim 40$ W m⁻², $\Phi_{ATM}^e \sim 60$ W m⁻²), while in smoke mist the increase in single scattering albedo
629 leads to almost halving of Φ_{ATM}^e . We note that this work presents estimates of daytime values of aerosol
630 radiation effects under different atmospheric situations. The issue of what is the uncertainty of these estimates,
631 caused by insufficiently exact information on the input parameters of radiation calculations, requires further
632 study.

633

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Table 1. Average (\pm SD) characteristics of AOD and water vapor content of the atmosphere in the period of smoke mist in 2012 as compared to multiyear data for July and “ordinary” smokes (April–October 2003–2011 and 2013)

Characteristics	July, multiyear		Smoke mist (Jun 17 – Aug 6, 2012)	“Ordinary” smokes
	Background conditions	Total dataset (with smokes)		
τ_{340}	0.25 \pm 0.11	0.33 \pm 0.26	1.37 \pm 1.10	0.58 \pm 0.28
τ_{500}	0.16 \pm 0.08	0.21 \pm 0.18	0.95 \pm 0.86	0.36 \pm 0.18
τ_{870}	0.07 \pm 0.04	0.09 \pm 0.08	0.40 \pm 0.40	0.15 \pm 0.09
α	1.48 \pm 0.29	1.49 \pm 0.28	1.58 \pm 0.21	1.59 \pm 0.22
β	0.058 \pm 0.037	0.074 \pm 0.064	0.33 \pm 0.35	0.12 \pm 0.071
W, g/cm ²	2.09 \pm 0.56	2.16 \pm 0.57	2.19 \pm 0.58	1.93 \pm 0.79

Table 2. Average (\pm SD), minimal, and maximal values of AOD (550 nm) in different periods of 2012 smoke mist according to data of ground-based and satellite measurements over the central part of the Western Siberia

Period	Satellite data		Ground-based observations (Tomsk-22)	
	Average \pm SD	min/max	Average \pm SD	min/max
June 6–10	0.23 \pm 0.08	0.04/0.41	0.19 \pm 0.07	0.10/0.27
July 1–4	1.40 \pm 0.62	0.18/3.25	1.51 \pm 1.05	0.85/2.72
July 10–14	0.28 \pm 0.11	0.08/0.76	0.22 \pm 0.11	0.11/0.35
July 24–28	1.19 \pm 1.08	0.00/3.63	2.74 \pm 1.07	1.32/3.83



Table 3. Optical and microphysical properties of biomass burning aerosol retrieved from AERONET Tomsk sites (“Tomsk”, “Tomsk-22”)

Aerosol characteristics	Smoke mist, 2012	Ordinary smoke, 2003-2011,2013
$n(440/675/870/1020)$	1.469/1.486/1.503/1.499	1.454/1.471/1.485/1.499
$\kappa(440/675/870/1020)$, ($\times 10^2$)	0.673/0.538/0.501/0.488	1.126/1.005/1.057/1.06
$\omega(440/675/870/1020)$	0.96/0.96/0.95/0.95	0.92/0.91/0.89/0.88
$g(440/675/870/1020)$	0.68/0.59/0.54/0.51	0.68/0.59/0.55/0.54
$r_v^f(\mu\text{m})$, $\sigma^f(\mu\text{m})$	0.181±0.02; 0.507±0.058 $r_v^f = 0.166 + 0.017\tau_{440}$ ($R=0.47$)	0.161±0.030; 0.422±0.058 $r_v^f = 0.153 + 0.011\tau_{440}$ ($R=0.13$)
$r_v^c(\mu\text{m})$, $\sigma^c(\mu\text{m})$	3.321±0.365; 0.703±0.056 $r_v^c = 2.928 + 0.44\tau_{440}$ ($R=0.66$)	2.911±0.665; 0.694±0.077 $r_v^c = 3.10 - 0.249\tau_{440}$ ($R=-0.13$)
$C_v^f(\mu\text{m}^3/\mu\text{m}^2)$	0.11±0.065 $C_v^f = 0.009 + 0.114\tau_{440}$ ($R=0.96$)	0.097±0.042 $C_v^f = 0.017 + 0.106\tau_{440}$ ($R=0.87$)
$C_v^c(\mu\text{m}^3/\mu\text{m}^2)$	0.025±0.012 $C_v^c = 0.013 + 0.014\tau_{440}$ ($R=0.62$)	0.062±0.04 $C_v^c = 0.036 + 0.035\tau_{440}$ ($R=0.24$)

Table 4. Parameters of radiation calculations

Case	τ_{550}	$\alpha_{440-870}$	W	ω_{550}	g_{550}
1 Background aerosol	0.13	1.46	2.1	0.925	0.7
2 “Ordinary” smokes	0.33	1.59	1.9	0.92	0.64
3 Smoke mist, 2012	0.84	1.58	2.2	0.96	0.64
4 July 14	0.34	1.74	2.5	0.98	0.64
5 July 27	3.54	1.54	1.2	0.94	0.65
Spectral surface albedo	$A_{s,470}=0.036$; $A_{s,560}=0.067$; $A_{s,670}=0.068$; $A_{s,870}=0.251$; $A_{s,1250}=0.285$; $A_{s,1650}=0.196$; $A_{s,2150}=0.1$				

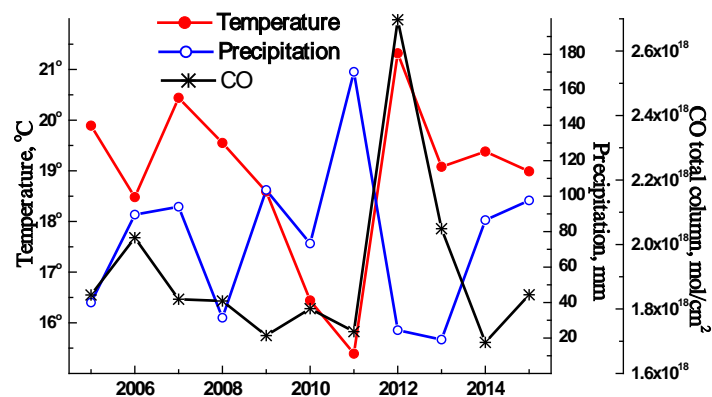


Figure 1. Interannual variations in July-average temperatures and precipitation amount according to data from “Tomsk” meteorological station, as well as interannual variations in total carbon monoxide content according to satellite observations over the territory of Tomsk region (<http://giovanni.sci.gsfc.nasa.gov/>).

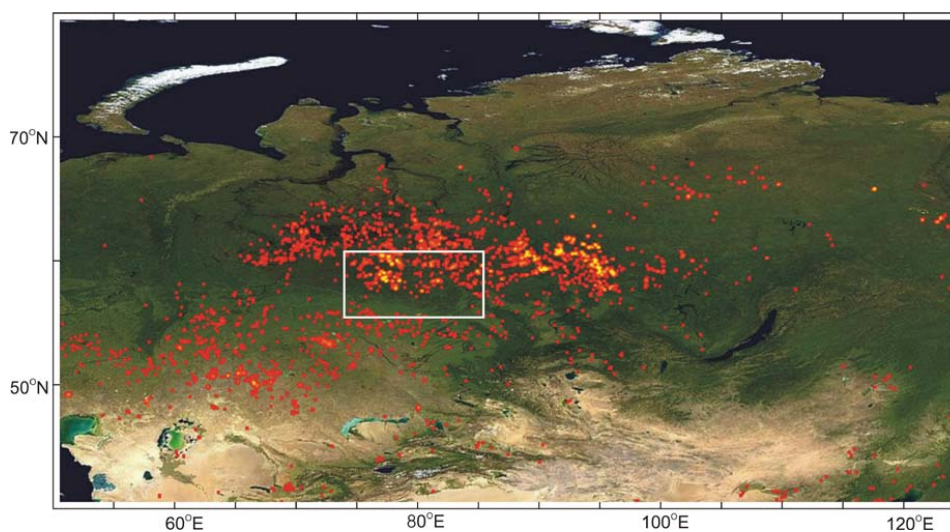


Figure 2. The map of forest fires over Siberia in the period from July 19 to 28, 2012 (<http://lance-modis.eosdis.nasa.gov/cgi-bin/imagery/firemaps.cgi>). The Tomsk region (56-61°N; 75-88°E) is highlighted by box.

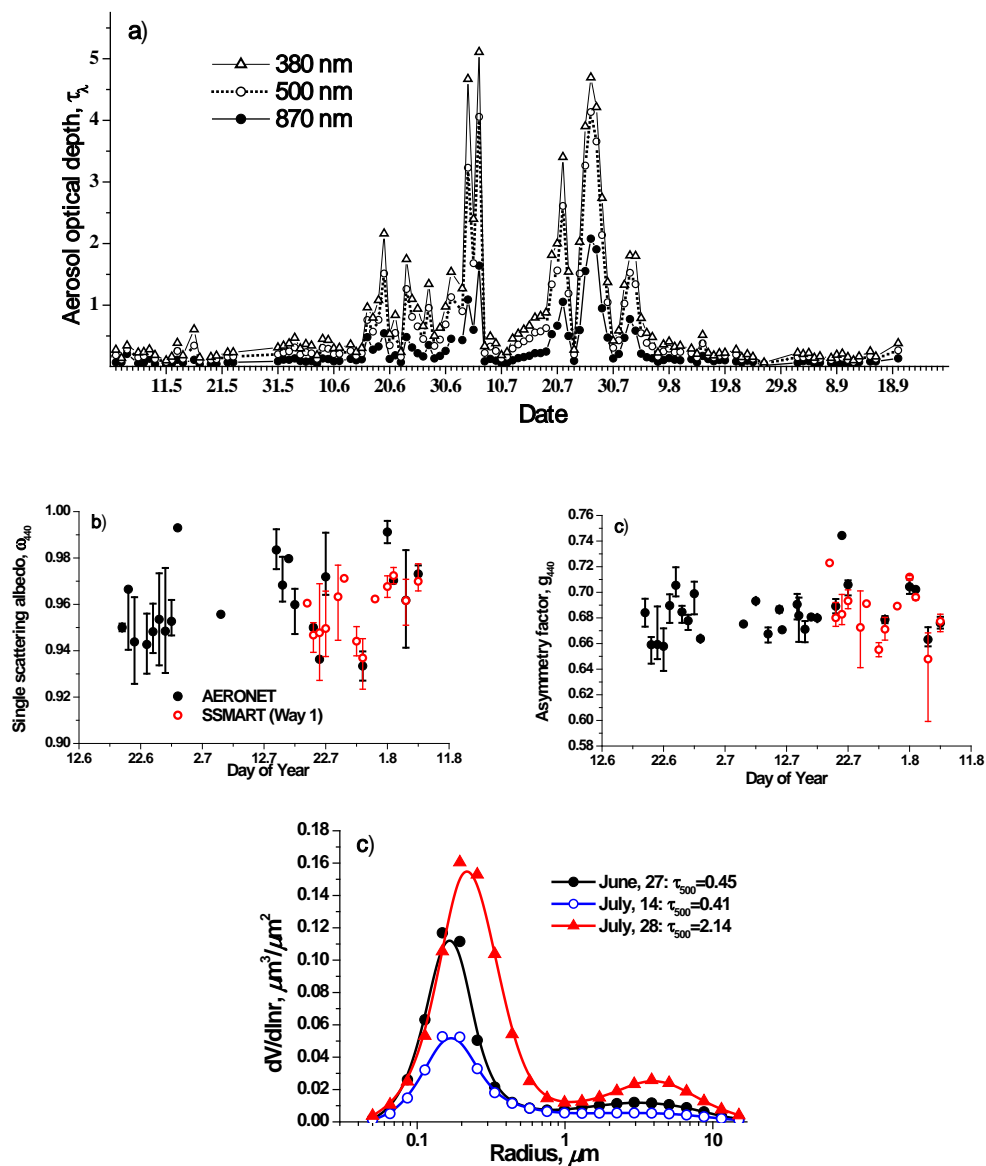


Figure 3. (a) Daily average AOD, (b) single scattering albedo and (c) asymmetry factor time series, and (d) examples of particle size distribution in “Tomsk-22” during summer period of 2012.

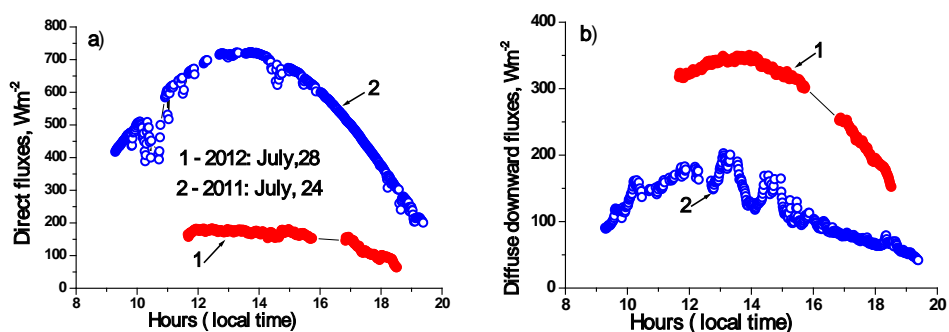


Figure 4. Daytime behavior of (a) direct and (b) diffuse fluxes of solar radiation under the usual conditions (July 24, 2011) and during smoke mist (July 28, 2012) in the region of “Fonovaya” observatory.

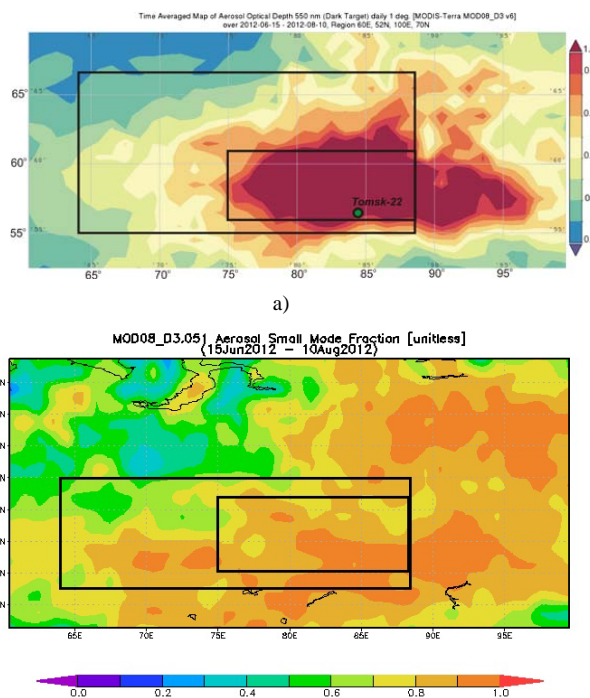


Figure 5. Spatial distributions of (a) AOD (MODIS collection 6) and (b) Aerosol Small Mode Fraction (MODIS collection 5) over Western and Eastern Siberia during June 15 – August 10, 2012. The central part of the Western Siberia (55-62°N; 64-88°E) and the Tomsk region (56-61°N; 75-88°E) are highlighted by boxes.

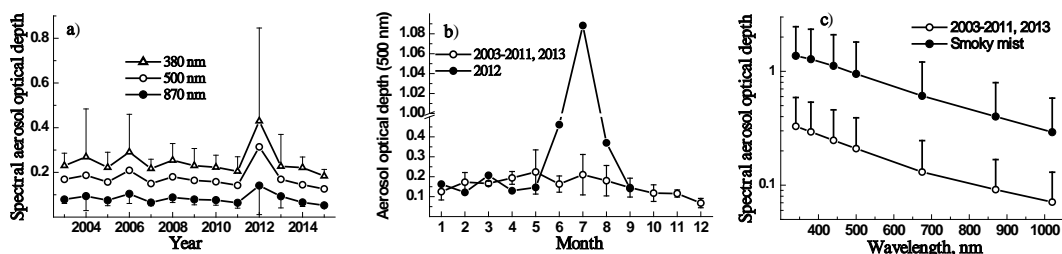


Figure 6. (a) Multiyear averages of spectral aerosol optical depth; (b) monthly averages of τ_{500} in 2012 compared with the multiyear averages for the period of 2003-2011, 2013; and (c) average spectral AOD dependences in the period of smoke mist as compared to multiyear data in July (Toms, 2003-2011, 2013, combined dataset of smoke and background situations)

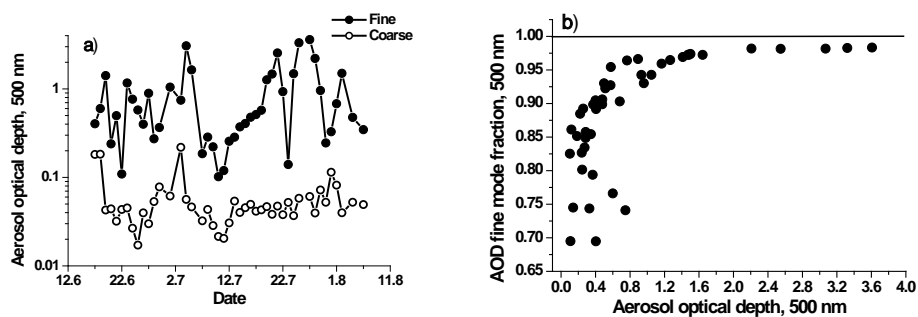


Figure 7. (a) Time series of computed average fine and coarse AOD mode; (b) fine mode fraction of AOD as a function of AOD (500 nm) for the Toms data during smoke mist in 2012

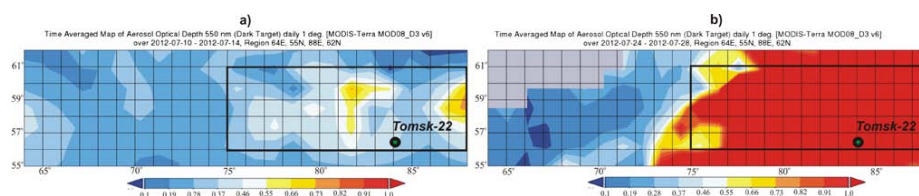


Figure 8. Spatial AOD distribution over the central part of the Western Siberia according to MODIS data for summer 2012: a) July 10-14, b) July 24-28.

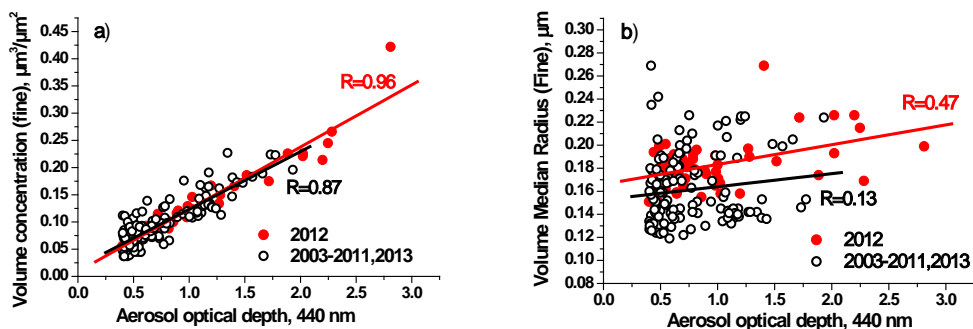


Figure 9. (a) Fine mode volume concentration and (b) median radius versus AOD(440 nm) in Tomsk.

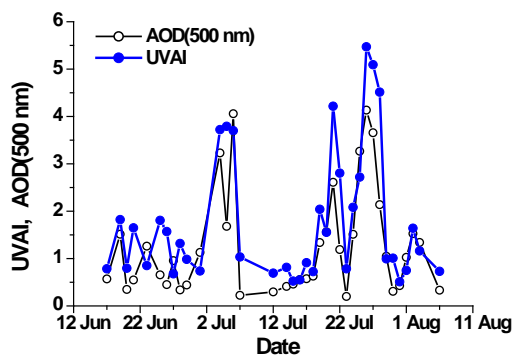


Figure 10. Ultraviolet Aerosol Index (<http://giovanni.sci.gsfc.nasa.gov/giovanni/>) near Tomsk (55-57°N, 83-85°E) and AOD according to data of ground-based measurements (<http://aeronet.gsfc.nasa.gov>)

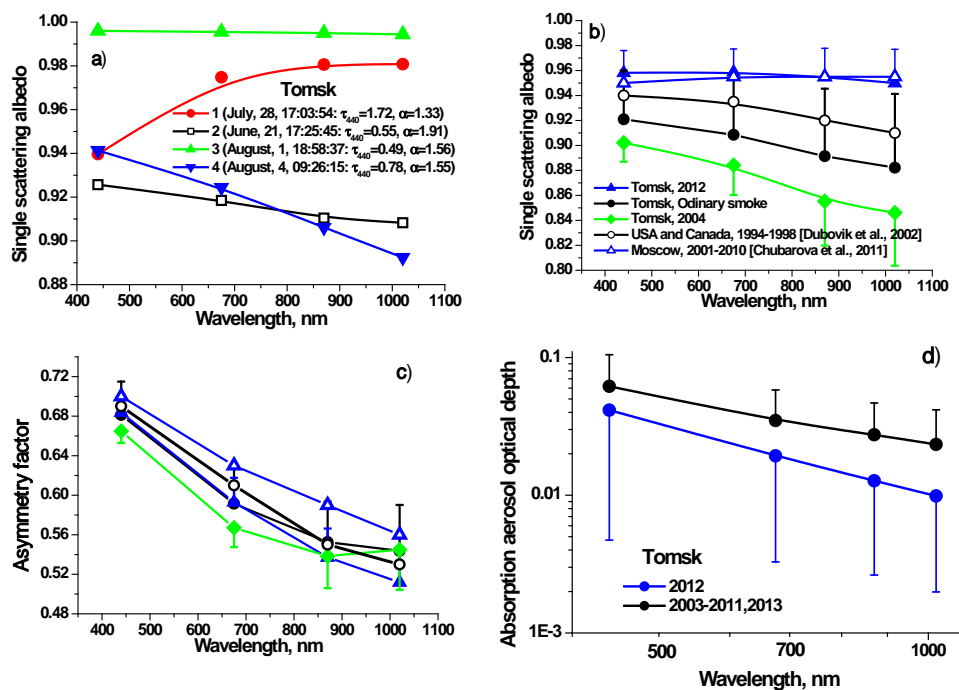


Figure 11. (a) Examples of spectral dependence of SSA; averaged (b) SSA and (c) AF for different periods and regions of observations; and (d) average spectral absorption aerosol optical depth for 2012 smoke mist and “ordinary” smokes.

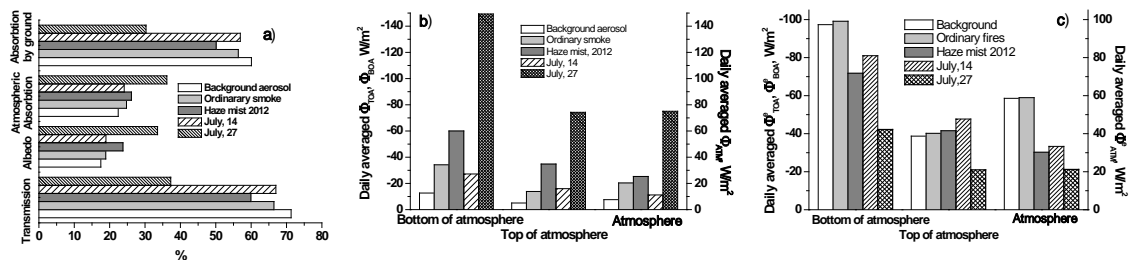


Figure 12. Radiative effects of aerosol under different atmospheric conditions.