

Chemical and dispersal characteristics of particulate emissions from forest fires in Siberia

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Abstract. Approximately 20 experimental fires were conducted on forest plots of 1–4 ha each in 2000–07 in two types of boreal forests in central Siberia, and 18 on 6 × 12-m plots in 2008–10. These experiments were designed to mimic wildfires under similar burning conditions. The fires were conducted in prescribed conditions including full documentation on pre-fire weather, pre-fire and post-fire forest fuels, fire intensities, and other biological, physical and chemical parameters. The amount of particulate matter emitted during a typical fire averaged 0.6 t ha⁻¹ and ranged within 0.2–1.0 t ha⁻¹ depending on burning conditions. Particulates accounted for ~1–7% of the total mass of the consumed biomass during a typical forest fire (10–30 t ha⁻¹ based on our data from 2000–07). Most of the particulate matter consists of organic substances, 77% on average, with a range of 70–90%. Elemental carbon averaged 8%, with a range of 2–18%. Trace element compositions and amounts of particulates indicate that there was no actual difference in the element emissions sampled from the fires conducted in the two forest types (6–8% in larch forest and 8% in pine forest). Most of the particulate matter, 90–95%, consists of submicrometre and near-micrometre particles ~0.1–5 µm in diameter.

Additional keywords: black carbon, chemical composition, elemental carbon, organic carbon, smoke particulate.

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Introduction

Burning of biomass is a natural phenomenon, with 3–6 × 10⁹ t of biomass burned annually worldwide. Although biomass burning and the accompanying chemical, atmospheric and weather and climate aspects relate to all regions, the focus of the present study is the wildfires burning in boreal forests of Siberia. The Russian boreal forests are estimated to cover ~700 × 10⁶ ha, with most of this in closed-canopy coniferous forests in Siberia. Satellite observations indicate that here wildfires burn an average of 10–14 × 10⁶ ha in forest, forest–steppe and non-forest types annually (Cahoon *et al.* 1994; Conard and Ivanova 1997; Sukhinin *et al.* 2004). Burning forest fuels yield large amounts of gaseous products and solid and liquid smoke particulates affecting radiation heat exchange in the air, aerosol and cloud interaction, and carbon emission and cycling (Wild 1999; Conard *et al.* 2002; Andreae *et al.* 2004; Soja *et al.* 2004).

The fraction of particulate emissions (smoke factor) represents only 1–7% of the biomass consumed under typical fire conditions (our results and reference data), but these particles can play a large role in the transfer of solar radiation through the atmosphere (Seinfeld and Pandis 2006). This is largely due to the presence of submicrometre particles, which contain a considerable proportion of elemental carbon (also called soot and black carbon, but sometimes these are not the same).

The submicrometre particles can remain in the atmosphere for a long time (generally 10–15 days), where they very effectively scatter and absorb sunlight and, thus, disturb the heat transfer between solar radiation, the atmosphere and the land. A postulated climatic effect of particulate emission (presumably, atmospheric cooling) could partly compensate for atmospheric warming presumably resulting from the increased emission of gaseous combustion products (carbon dioxide and methane). Besides, emission of large amounts of fire particulates followed by the long-distance dispersion of smoke matter in the atmosphere worsens respiratory quality of the near-surface air over a huge area. Consequently, quantitative data on smoke production are required to predict the chemical and optical changes occurring in the atmosphere from seasonal forest fires. These quantitative data are critical for computational modelling of regional and global weather and climate changes, and for quantitative or qualitative predicting respiratory and toxic properties of the near-surface air. It should be noted that the hypothetical weather and climate effect should directly depend on the smoke emission properties and the altitude of smoke plumes, and indirectly on the post-fire changes of snow and ice albedo, additionally influencing the radiation budget over a territory polluted with smoke deposits (Betts and Ball 1997; Betts 2000; French 2002).

This paper reports on the physical and chemical characteristics of particulate emissions sampled from a series of experimental fires conducted from 2000 to 2010 in boreal forests of central Siberia. Some results from the earlier fire experiments conducted in Scots pine (*Pinus sylvestris*) forests in 2000–03 have already been published (Samsonov *et al.* 2005; McRae *et al.* 2006). Later experiments were conducted in larch (*Larix* sp.) mixedwood forest (2006–07) and pine forest (2008–10) where other trees were present too. The various forest types, ~500 km remote from each other, represent a range of soil, vegetation, landscape and climatic conditions. In this regard, the analysis completed by the present study should be useful for quantifying and comparing mass concentrations, smoke yields (smoke factors), chemical composition, dispersal and morphological characteristics of particulate emissions from fires burning in these different forest types.

Materials and methods

Study area and field experimental design

The fire experiments were conducted at four research sites located near the middle parts of the Yenisey River (Yartsevo site, 2000–02, pine forest) and the Angara River (Govorkovo 2002, Khrebtovo 2003, pine forest; Nevonka 2006–07, mixed larch forest). Fire plots measured ~200 × 200 m (11 plots of 4 ha each) except at Govorkovo, where the two plots were 2.3 and 3.1 ha, and Nevonka (five plots ~100 × 100 m of 1 ha). All plots were burned using line ignition along the windward side to create equilibrium fire behaviour that simulated wildfires under similar burning conditions (McRae *et al.* 2006). It should be noted that in most cases the model fires were ground fires of low or moderate intensity with fire spread rates of 1–3 m min⁻¹. Before and after each fire, a full inventory of pre-burn and post-burn fuels was performed both quantitatively and qualitatively. During the fire, qualitative observations and quantitative measurements were made of fire behaviour: rates of flame spread, fireline intensities, temperature distributions, etc. (the methodology of fire experiments and typical parameters of fires are described in McRae *et al.* (2006) and Samsonov *et al.* (2005)). An important advantage of using experimental fire plots is the comprehensiveness of the study under controlled conditions where full documentation of all processes can be completed.

In 2008–10, 18 fires were additionally conducted at a pine forest (Govorkovo site) using small plots 6 m wide and 12 m long. These plots were covered with a plastic film at a height of 0.5–1 m over the plots to protect them from rain. Fires at the 6 × 12-m plots produce (i.e. mimic) smoke matter that is similar to smoke emissions (in both the chemical, dispersal and morphological properties) from large forest fires.

Aerosol filters and sampling procedure

The sampling of particulates was carried out by pumping smoke through circular 70-mm polymeric fine-fibre filters (Analytical Filter for Aerosols – Chemical Analysis, the AFACHA filter, Russia) and 50-mm glass-fibre filters (Gelman Science, Ann Arbor, MI). Before use, all filters were carefully dried and weighed under laboratory conditions. After the fires, the filters were weighed using the same procedure. The mass concentrations of smoke particles were calculated based on the increase in

weights of the filters, pump flow rates and pumping duration, which were recorded for each sampling. The flow rate and the time length were respectively 130 L min⁻¹ and 3–6 min in the case of polymeric filters, and 30 L min⁻¹ and 7–15 min for the glass-fibre filters. During a typical burn period of 1–3 h for the 1–4-ha plots (~30 min for the 6 × 12-m plots), four to eight smoke samples were taken on the Gelman filters and six to twelve probes using the AFACHA filters. Smoke samples were taken at different positions on the leeward outer side of the fire plot, but the sampling point was periodically shifted as the fire front progressed down the plot. The filter holders were located at a height of 0.5–1.0 m over burning and smoking fuels (moss, lichen, shrubs, fallen wood). At this position, the primary hot particles, 40–50°C but sometimes hotter, were sampled. These freshly formed hot particulates had no time to react with atmospheric moisture and be subjected to any post-fire chemical reactions and physicochemical conversions in the air.

Chemical analysis of particulate emission

The AFACHA filters were used to measure the quantities of trace elements in smoke particulates. Synchrotron radiation X-ray fluorescence (SRXRF) (Baryshev *et al.* 1995; Chankina *et al.* 2001) was applied to detect 20 chemical elements from potassium (K) to lead (Pb). The SRXRF sensitivities were earlier determined by Koval'skaya (2001) and Baryshev *et al.* (2002). As applied to the AFACHA samples, the detection limits of surface concentrations ranged from 0.05–0.1 µg cm⁻² for calcium (Ca) and K to 0.0004–0.001 µg cm⁻² for strontium (Sr), zirconium (Zr) and molybdenum (Mo).

The amounts of carbonaceous matter sampled on glass-fibre filters were measured by reaction gas chromatography using a facility described in Popova *et al.* (2004, 2007). This method (also called 'the simple thermal method') is based on catalytic oxidation of organic matter into carbon dioxide followed by catalytic conversion into methane. In calculating the total mass of organic matter, one must take into account that the carbonaceous molecules also contain atoms of oxygen, hydrogen and nitrogen. The organic constituents of smoke particles consist of the chemical products formed from thermally decomposing biomass consisting of cellulose, lignin, waxes and tars. The mass fraction of carbon atoms in these compounds ranges from 44 to 55%, with a 50% average usually being cited in most papers (Konev 1977; Levine and Cofer 2000; Levine *et al.* 2000). Therefore, the carbon mass measured by the above equipment should be doubled to determine the total organic mass in samples. The coefficient used in our earlier papers was 2.00 (stearic acid, oleic acid and benzo(a)pyrene were used as the carbonaceous standard masses, giving coefficients of 1.98–2.13), resulting, however, in a systematic 10–15% underestimation of organic matter found in these samples. Later, we began to use dry pine wood as an additional standardised organic mass, and the coefficient was found to be 2.20–2.25. This value is used in the present paper to determine the total mass of organic constituents in samples taken in 2006–10. The above chromatographic method was applied to measure elemental carbon too. The elemental carbon on a glass-fibre filter burned with oxygen to form CO₂. Other steps in the procedure were similar to those described for the determination of organic carbon. Pure graphitic

carbon was used as the calibration standard to determine the amounts of elemental carbon in samples.

Measurement of dispersal characteristics of smoke emission

A rotary impactor and a five-cascade inertial impactor were used to measure the size distributions of smoke particulates. The rotary impactor consists of an electric motor that rotates an aluminium rod with blades 100 mm long at a rate of 1500 cycles min^{-1} . Two glass plates, 5 mm wide and 25 mm long, are fixed at the ends of each blade. The plates are coated with a thin layer of viscous matter (Apiezon M grease) to provide a sticky surface for particles to adhere to after impact. The impactor was positioned in the fire smoke for 15–60 s. Particles with sizes greater than $\sim 10 \mu\text{m}$ can impinge effectively on the plates. Sampling of particles of less than 3–5 μm is not practical with this device. The plates with particles were photographed with a digital Axioscope 2 microscope (Carl Zeiss, Jena, Germany). The images of particles were analysed using *MapInfo* software (MapInfo Corp., Troy, NY, USA) to determine the size parameters of individual particles (cross-section diameters, total perimeters and cross-section areas) and to calculate the average characteristics for the group of particles (mean size, standard geometric deviation).

The impaction samplings of smoke particulates were carried out in the model fires in 2007–10 using the five-cascade impactor (designed and produced in the Institute of Chemical Kinetics and Combustion, Russia). The operation principle is based on an abrupt 90° turn of an ‘air-and-smoke’ stream for each cascade followed by the deposition of particles of a certain size range on this cascade. The operating characteristics are as follows: the particles of aerodynamic diameters more than $33\text{-}\mu\text{m}$ deposit on the first cascade, the particles ranging within $33\text{--}19 \mu\text{m}$ deposit on the second cascade, the $19\text{--}7.5\text{-}\mu\text{m}$ particles on the third cascade and $7.5\text{--}3 \mu\text{m}$ on the fourth. These cascades are the glass plates. The plates are coated with sticky grease, dried and weighed. After the experiments, they are dried and weighed to determine the amounts of particulates of the various size ranges. The fifth cascade is a couple of AFACHA filters, which collect the particles $3\text{--}0.1 \mu\text{m}$ in diameter. After the fire, these filters are dried and weighed to determine the mass of fine particulates of less than $3 \mu\text{m}$. The impaction of smoke emission was performed twice for every fire experiment (37 impactions of 15–20 min length in 19 fires; in total, 148 glass plates with deposited smoke particulates and 74 filters from the fifth cascade).

An optical counter (analyser) of aerosol particles PKZV-906 (Inform-Pribor, Moscow, Russia) was used *in situ* to measure the dispersal characteristics of fine particulates ranging from $0.2\text{--}0.3$ to $5\text{--}8 \mu\text{m}$. The smoke was pumped from zones of either flaming, smouldering or mixed combustion. A 20:1 flow of pure air was added to cool the hot smoke before it entered the counter (more than 400 measurements of 1 min each in 19 fires).

Results and discussion

Mass concentrations and smoke yields (smoke factors) from fires in pine and larch forests

The mass concentrations of smoke particulates measured in the fires conducted on either the pine forest or the larch sites were found to be almost similar. This is based on the average of 13

fires in pine forest, resulting in a total average of 46 mg m^{-3} (Samsonov *et al.* 2005). The total average found for five fires in larch forest was 50 mg m^{-3} (41, 36, 29, 25, 56, 76, 70, 64, 47 and 60 mg m^{-3} ; Tables 1 and 2; two sets of measurements of smoke concentration were performed in each fire using the polymeric and glass-fibre filters). This is evidence that the mass emission processes are weakly dependent on the kinds of biomass. On our forest plots, the total amounts of fuel ($41\text{--}62 \text{ t ha}^{-1}$ on the pine sites and $49\text{--}78 \text{ t ha}^{-1}$ on the larch sites), and the species diversity and morphological composition of forest fuels were quite similar. Mainly, the combustion and emission rates depended on the duration of dry pre-fire weather and on the burning conditions during the fire, and these were quite similar for our fires.

A specific characteristic of forest fires is that the concentrations measured during a certain fire but at different sampling positions were found to differ greatly from each other (Tables 1, 2). These variations are due to both the non-uniform distribution of fuels on a plot and the irregular changes in wind force and direction during the fire. The changeable burning due to both fuel-bed spatial heterogeneity (including different dryness factors of fuels) and the fluctuation in wind conditions results in irregular changes of smoke concentrations. These positional concentrations were averaged for each fire and then for all fires to obtain the above values of 46 and 50 mg m^{-3} , which characterise the smoke capacity of the fires under study. In fact, the positional concentrations presented in the tables range too widely (e.g. 6 and 141 mg m^{-3} in Table 1) and they are systematically dependent on the burn efficiency of fuels and the physical parameters of combustion (flaming or smouldering or mixed combustion) to suppose a normal (Gaussian) distribution law.

There was almost vertical movement of hot smoke of velocity $U = \sim 0.3\text{--}1.5 \text{ m s}^{-1}$ at a sampling height of $0.5\text{--}1.0 \text{ m}$. If the smoking duration $t = \sim 5\text{--}40 \text{ min}$ and if the mass concentration C (mg m^{-3}) is determined, we can calculate the total quantity of particulate matter M (t ha^{-1}) emitted from 1 ha of the burned forest by using the following equation (the factor 0.0006 is due to the units used):

$$M = 0.0006C \times U \times t \quad (1)$$

From the average concentration $C = \sim 48 \text{ mg m}^{-3}$, $U = \sim 1 \text{ m s}^{-1}$ and smoking time $t = \sim 20 \text{ min}$, we calculated the average value for $M = \sim 0.6 \text{ t ha}^{-1}$, but it ranges from 0.2 to 1.0 t ha^{-1} depending on burning conditions. Thus, the particulate yield (smoke factor) was $\sim 1\text{--}7\%$ of the total mass of the biomass consumed during a typical forest fire ($10\text{--}30 \text{ t ha}^{-1}$ based on our data from 2000–07). At sampling heights of $2\text{--}3 \text{ m}$ and higher, the smoke began to bend over owing to wind conditions and mixing with ambient air. Consequently, smoke measurements at such heights can characterise the smoke concentrations at these points, but it is impossible to derive a relevant value for M from Eqn 1 for any point other than at the height of $0.5\text{--}1.0 \text{ m}$ where the smoke rises almost vertically.

Contents of trace elements in smoke emission and their origins

In filter samples from our earlier pine-forest fires, the amounts of trace elements (K, Ca, Ti, Fe and another 15–16 elements)

Table 1. Trace elements found in smoke samples taken from fires burning in larch mixedwoods of central Siberia

Concentrations are presented in the volume units ($\mu\text{g m}^{-3}$), but they were calculated from surface concentrations ($\mu\text{g cm}^{-2}$) determined by the synchrotron radiation X-ray fluorescence (SRXRF) method

Concentration of trace elements in smoke ($\mu\text{g m}^{-3}$)																					
K	Ca	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	As	Br	Rb	Sr	Zr	Nb	Mo	Pb	Total mass concentration of particulate matter (mg m^{-3})	Estimate of percentage of soil–mineral matter (%)
Fire experiment of 10 July 2007																					
222	0	1.8	0.33	0.33	0.7	4.3	0.033	0.133	0.200	0.333	0.000	0.000	0.033	0.033	0.017	0.050	0.000	0.018	0.100	6	5
65	67	6.7	0.33	0.83	4.3	15.7	0.000	0.067	0.100	0.167	0.017	0.000	0.183	0.100	0.267	0.067	0.017	0.007	0.050	19	9
243	28	1.7	0.00	0.50	1.5	3.7	0.067	0.017	0.183	0.167	0.050	0.000	0.167	0.050	0.067	0.017	0.000	0.023	0.050	25	4
220	147	4.3	0.33	1.17	16.3	11.0	0.067	0.100	0.233	1.500	0.050	0.000	0.700	0.250	0.483	0.083	0.017	0.005	0.150	141	3
85	63	9.0	0.00	0.67	4.8	27.5	0.083	0.050	0.167	0.500	0.017	0.067	0.300	0.150	0.217	0.067	0.000	0.027	0.050	72	3
117	112	4.7	0.00	0.17	6.0	16.5	0.050	0.117	0.333	0.667	0.050	0.117	0.267	0.083	0.367	0.017	0.000	0.000	0.000	65	4
100	287	11.5	1.83	0.50	10.7	20.8	0.017	0.150	0.483	0.667	0.033	0.183	0.233	0.117	1.083	0.067	0.000	0.017	0.000	55	9
100	170	7.7	1.50	1.00	16.7	13.8	0.083	0.200	0.267	0.667	0.033	0.050	0.450	0.117	0.467	0.067	0.000	0.013	0.067	96	3
37	22	6.2	0.33	0.33	1.2	9.5	0.117	0.050	0.050	0.500	0.030	0.030	0.070	0.070	0.130	0.05	0.000	0.010	0.000	25	4
Mean value:																					
5																					
Fire experiment of 15 July 2007																					
118	52	2.2	0.33	0.83	3.5	7.7	0.000	0.000	0.150	1.167	0.033	0.000	0.850	0.133	0.083	0.017	0.000	0.022	0.167	11	18
93	72	2.5	0.50	0.33	4.0	5.8	0.117	0.017	0.050	0.167	0.017	0.000	0.367	0.067	0.083	0.000	0.017	0.000	0.117	61	3
478	93	6.5	0.00	0.50	4.8	10.8	0.000	0.133	0.733	3.500	0.033	0.083	1.467	0.383	0.067	0.067	0.017	0.000	0.233	57	11
103	98	4.2	0.00	1.33	3.7	11.2	0.150	0.267	0.750	1.333	0.050	0.000	0.567	0.117	0.100	0.050	0.017	0.015	0.200	48	5
117	223	4.7	0.50	0.50	11.7	11.7	0.000	0.050	0.167	1.667	0.000	0.000	0.433	0.100	0.333	0.050	0.017	0.028	0.100	51	7
140	97	13.5	1.33	0.00	8.5	23.0	0.050	0.117	0.633	1.000	0.050	0.000	0.300	0.150	0.133	0.083	0.017	0.000	0.217	46	6
242	418	22.7	8.00	0.33	12.3	12.5	0.000	0.183	1.833	2.833	0.033	0.033	0.433	0.117	4.383	0.100	0.033	0.038	0.167	133	19
80	267	2.7	0.50	0.83	12.8	22.0	0.233	0.267	1.050	7.833	0.050	0.000	0.450	0.067	0.433	0.083	0.017	0.020	0.350	76	5
142	58	5.3	0.33	0.17	3.7	9.7	0.050	0.000	0.100	3.500	0.033	0.000	0.967	0.217	0.133	0.083	0.017	0.000	0.067	14	16
60	7	0.7	0.33	0.00	1.5	0.0	0.000	0.000	0.000	0.667	0.000	0.000	0.467	0.050	0.000	0.000	0.000	0.008	0.167	6	11
88	95	3.2	0.83	0.17	4.2	5.3	0.000	0.100	0.817	2.667	0.033	0.000	0.750	0.133	0.050	0.017	0.000	0.005	0.150	12	17
Mean value:																					
11																					
Fire experiment of 17 July 2007																					
138	38	23.7	0.67	1.00	3.8	41.7	0.167	0.117	0.183	0.333	0.033	0.000	0.167	0.167	0.150	0.183	0.050	0.023	0.050	24	10
400	1467	18.8	3.00	0.33	113.3	53.3	0.267	0.150	0.650	5.667	0.117	0.000	0.383	0.183	1.717	0.150	0.017	0.007	0.133	117	18
270	288	3.8	0.00	0.67	18.3	4.8	0.000	0.100	0.217	2.667	0.017	0.000	0.200	0.100	0.283	0.000	0.017	0.013	0.017	44	13
1115	112	3.3	0.67	0.33	7.0	0.0	0.033	0.017	0.100	0.833	0.000	0.000	0.117	0.033	0.083	0.017	0.000	0.015	0.017	41	6
198	447	9.5	1.17	0.17	41.7	18.3	0.000	0.050	0.167	2.333	0.033	0.000	0.133	0.083	0.517	0.067	0.000	0.000	0.150	64	11
158	130	0.7	0.00	0.67	6.0	0.0	0.050	0.000	0.217	0.333	0.083	0.083	0.017	0.033	0.117	0.033	0.000	0.000	0.000	170	2
1115	227	4.0	0.50	0.17	16.7	5.5	0.000	0.000	0.117	0.667	0.033	0.067	0.033	0.050	0.250	0.050	0.000	0.042	0.000	22	17
210	660	21.7	2.67	0.17	43.3	41.7	0.133	0.100	0.300	2.333	0.000	0.000	0.800	0.250	0.567	0.117	0.000	0.000	0.433	44	22
163	137	10.2	1.00	0.67	11.7	15.7	0.083	0.033	0.050	0.333	0.117	0.000	0.033	0.083	0.150	0.050	0.017	0.005	0.067	108	3
Mean value:																					
70																					

Table 2. Contents of carbonaceous matter in smoke samples taken from fires burning in larch mixedwoods of central Siberia

Sample weight (mg)	Total mass concentration (mg m ⁻³)	Measured organic matter in sample		Measured elemental carbon in sample		Total share of carbonaceous substance (%)	Filter's exterior appearance
		(mg)	(%)	(mg)	(%)		
Fire experiment of 23 July 2006							
11.8	39	8.4	71	0.57	5	76	—
9.9	30	7.6	77	0.53	5	82	—
6.1	31	6.7	110	0.37	6	116	—
8.3	42	8.4	101	0.60	7	108	—
Mean:	36		90		6	96	
Fire experiment of 3 August 2006							
3.7	19	2.4	65	0.68	18	83	—
7.2	24	4.9	68	0.26	4	72	—
6.7	22	4.4	66	0.22	3	69	—
5.1	17	4.4	86	0.17	3	89	—
3.8	13	3.5	92	0.08	2	94	—
11.6	37	7.6	66	0.96	8	94	—
Mean:	25		74		6	80	—
Fire experiment of 10 July 2007							
1.9	16	1.2	64	0.2	10	74	Black
40.0	133	26.0	65	3.4	9	74	Yellow-brown, oily
16.3	54	11.4	70	1.8	11	81	Black
30.6	102	25.0	82	3.7	12	94	Brown, oily
Mean:	76		70		11	81	
Fire experiment of 15 July 2007							
19.1	64	13.2	69	1.5	8	77	Yellow-brown
18.6	62	17.5	94	1.9	10	104	Brown, oily
21.9	73	21.5	98	2.7	12	110	Brown, oily
9.8	33	7.1	72	0.7	7	79	Brown, oily
43.7	146	31.9	73	2.9	7	80	Brown, oily
6.1	20	4.3	71	0.8	13	84	Black
24.1	80	15.7	65	1.3	6	71	Black
11.4	38	9.9	87	1.2	11	98	Black
Mean:	64		79		9	88	
Fire experiment of 17 July 2007							
7.4	25	5.4	73	0.4	6	79	Black
10.9	36	6.7	62	0.6	6	68	Yellow-brown
15.5	52	14.3	92	1.4	9	101	Black
26.6	89	26.6	100	1.7	7	107	Yellow-brown, oily
12.4	41	6.5	53	0.6	5	58	Black
11.4	38	6.4	56	0.9	8	64	Black
26.1	131	15.2	58	1.7	7	65	Brown
16.8	84	20.8	124	1.0	6	130	Yellow-brown
9.2	46	4.9	53	0.8	8	61	Black
Mean:	60		75		7	82	

were determined by the SRXRF method. These elements exist in the form of chemical salts, oxides and complex compounds, which form soil matter. The main contribution, more than 95%, was made up of six elements: K, Ca, Fe, Mn, Ti and Zn (Samsonov *et al.* 2005). The total mass of these and other 14–16 minor elements was 0.3–1% of the total mass of smoke aerosols. It should be noted, however, that SRXRF is insensitive to the 'light' elements (Na, Al, Si, O, N and C), and it has low sensitivity to P, S and Cl. Moreover, the soil matter emitted into the air consists for the most part of these undetectable elements (e.g. SiO₂, Al₂O₃), sometimes in concentrations 10-fold and more than the above detectable elements (Mason 1966; Rahn 1976; Perelmann 1979). Taking this into account, the above

percentage of trace elements, 0.3–1% determined by the SRXRF method, should be then multiplied up 10–20 times, resulting in 3–17% (~8% average) of the total soil–mineral mass emitted from our fires in pine forests (Samsonov *et al.* 2005).

Fires in larch forest gave similar results. The elements found in the emissions from three fires in 2007 demonstrate that the total fraction of soil–mineral matter is in a range 3–19%, with an average of 9% (in Table 1, the concentrations are in volume units calculated from the surface concentrations on filters determined by SRXRF). The data on two fires of 2006 are omitted in Table 1 but the calculated average was 13%. The main contribution, more than 95%, is made up of five elements, K, Ca, Fe, Ti and Mn. The so-called 'enrichment factor' can be used to

Table 3. Contents of trace elements in fine smoke aerosols and their main contributing sources for fires in larch mixedwood forests of central Siberia
Concentrations of all elements are normalised to the mean concentration of potassium K and, consequently, are expressed in dimensionless units (the normalised concentration of potassium K is assigned to be 100 dimensionless units)

Sample technique	Trace elements																			
	K	Ca	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	As	Br	Rb	Sr	Zr	Nb	Mo	Pb
Fifth impaction cascade	100	31	0.6	0.0	0.06	1.9	1.5	0.0	0.0	0.78	1.3	0.006	0.038	0.281	0.044	0.031	0.013	0.0	0.0	0.013
Direct sampling	100	149	5.6	0.5	0.28	10.9	9.3	0.03	0.03	0.10	0.7	0.023	0.010	0.108	0.057	0.156	0.038	0.001	0.007	0.053
Fifth cascade/direct sampling ratio	1.0	0.21	0.11	0.0	0.21	0.17	0.16	0.0	0.0	7.8	1.9	0.26	3.8	2.6	0.77	0.20	0.34	0.00	0.00	0.25
Main source of element	Plant	Soil–plant	Soil	Soil	Soil	Soil	Soil	Soil	Soil	Plant	Plant	Soil	Plant	Plant	Soil	Soil	Soil	Soil	Soil	Soil

discriminate between ‘soil’ and ‘plant’ origins of the emitted elements (Mason 1966; Rahn 1976; Samsonov *et al.* 2005). Certain elements, K, Br, Zn and some others, are known to be included in living tissues as they play a biological role in plant cells. During the thermal destruction and combustion of forest vegetation, these ‘vital’ elements should be released and incorporated in fine particles rather than in coarse ones. Iron, titanium, copper and some other elements originate mainly from soil and dust particles. This results in higher concentrations of ‘soil’ elements in the coarse particulates. Calcium (CaCO_3) originates from both the soil and dust particulates and plant tissues (see *Morphological properties and specificity of trace element composition of fine particulates* Section).

To correct the above estimated total shares of mineral matters in particulate emissions, one must take into account that the aforementioned 10-fold and more excess of Na, Si, Al and other ‘light’ elements relates only to the substances that originate from burning of the dust-laden forest litter and vegetation (soil and dust particles, settled on the forest floor and vegetation for many months and years before a fire, can re-enter the air during the fire). Assuming that only half of the calcium in fire smokes originates from soil matter, the above calculated 13 and 9% share of mineral matter in smoke sampled from fires of 2006–07 seems to be too high, but a value of 6–8% should be considered as more accurate. The element concentrations shown in Table 1 and the earlier paper (Samsonov *et al.* 2005) indicate that there was no actual difference in the soil–mineral emissions sampled from the fires conducted in two forest types, 6–8% in larch forest and 8–10% in pine forest.

Based on the average percentage of mineral substances of 8%, and from the yield of particulate matter of $0.2\text{--}1\text{ t ha}^{-1}$, we calculated the amount of mineral matter, $16\text{--}80\text{ kg ha}^{-1}$, emitted from 1 ha of burned forest and, in total, $\sim 600\,000\text{ t}$ annually from fires in the Asian part of Russia. However, these values characterise smoke emission sampled just over the burning plot. In terms of subsequent wind transport and redistribution of particulates in the atmosphere, a considerable portion of soil matter consisting of coarse particles of several tens of micrometres in diameter can deposit back onto land not far from the fire sites. In contrast, mineral substances originating from combustion of plant tissues included in the fine particles can be transported by wind many hundreds and thousands of kilometres. These long-lived elements are potassium, zinc, bromine, calcium to some degree and other biologically functional

elements (additional data on element concentrations are given in the following section and Table 3).

Carbonaceous substances in smoke emissions

Table 2 summarises the data on the amounts of carbonaceous matter sampled from fires at larch forest sites in 2006–07. Carbonaceous matter exists in particulates as a mix of organic compounds and elemental carbon. As follows from Table 2, the greatest proportion of particulate matter, $\sim 77\%$ on average, with a range of 70–90%, is the organic substances. Elemental carbon averages $\sim 8\%$, with a range of 2–18%. These data are in agreement with our earlier data (Samsonov *et al.* 2005) gathered from fires burning in pine forests (7–15% of elemental carbon, 50–70% of organic matter but, as is stated in *Materials and methods*, there was a 10–15% systematic underestimation of organic matter in the earlier measurements). Thus, mass concentrations, trace element composition and carbonaceous constituents of smoke emissions were practically the same for all of our fires regardless of forest type. As was pointed out in the previous sections, the key factor for fire behaviour is the pre-fire weather conditions (dryness factor). These were quite similar for the fires in the pine and larch forests. A similar conclusion that the gaseous and particulate smoke compositions strongly depend on the fuel consumption (i.e. on the weather effect on fuel dryness) and the physical parameters of combustion (flaming or smouldering combustion) and, to a lesser extent, on the vegetation type burned, is known from the literature (Ward *et al.* 1996; Cofer *et al.* 1997; Novakov *et al.* 1997). However, the vegetation types and combustion regimes have an evident effect on the dispersal and morphological properties of smoke particulates (following Sections).

Dispersal characteristics of smoke particulates

The dispersal parameters and the chemical composition of fine particles of submicrometre and $1\text{-}\mu\text{m}$ classes are of particular interest owing to the strong dependence of scattering and absorption of solar radiation on the size and chemical composition of particles. The submicrometre particles possess a high ability to scatter sunlight, but the greatest absorption is shown by the particles composed of carbonised materials (soot, black carbon). The lifetimes of particles suspended in the air depend on the particle’s size and smoke-plume heights. The smoke from our fires reached altitudes of 0.5–0.7 km (from observations

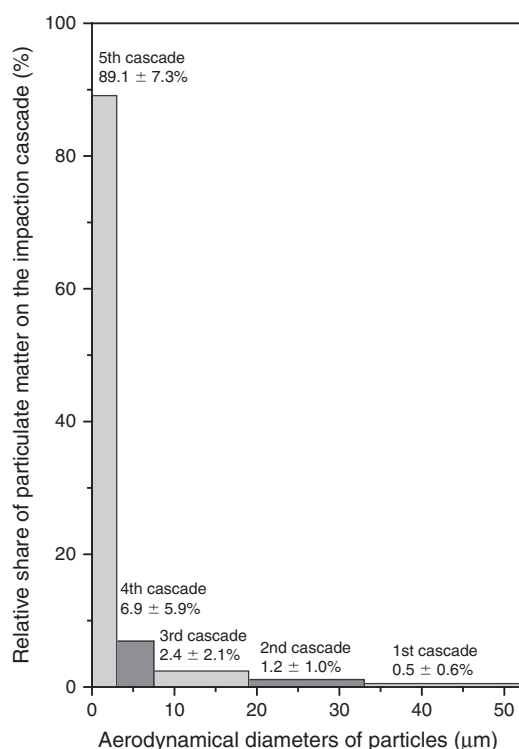


Fig. 1. Histogram of mass distribution of smoke emissions measured with a five-cascade impactor (by averaging the data of 37 impaction samplings from 19 model fires).

made by a helicopter used to monitor our fires; McRae *et al.* 2005), but the plume can be higher in large fires (Goldammer 1996). The submicrometre and 1–3- μm particles remain aloft in the air for 10–15 days (by that time, the particles are scavenged by precipitation, so-called ‘wet deposition’) and can be transported by wind over thousands of kilometres. The 30–100- μm particles fall to the ground in a few hours within 5–100 km from a fire site depending on local wind conditions. However, the mass of a particle is proportional to the particle diameter cubed, i.e. the weight of a 5- μm particle could exceed the total weight of several hundreds of submicrometre particles. Thus, to characterise fire emissions, one should determine both the size distribution of number concentrations of submicrometre particles (affecting light scattering and global transport) and the total mass distribution (affecting the chemical and respiratory properties of the near-land-surface air).

Fig. 1 presents a bar chart of the mass distribution of smoke matter sampled with a five-cascade impactor (37 samplings of 15–20 min each from 19 model fires including the 100 × 100-m fires in 2007). We see that the major portion of emission, ~90%, is composed of fine particles of less than 3 μm , and ~7% of particles 3–7 μm thick. The particulates larger than 7 μm contribute only 2–5% of the total smoke matter. It should be noted, however, that it was practically impossible for coarse smoke particles of 30–50 μm and larger to enter the impactor’s inlet and reach the first impaction plate without loss at the inlet walls. In addition, ash particulates several tens and hundreds of micrometres long, composed of solid mineral matter (K_2CO_3 and

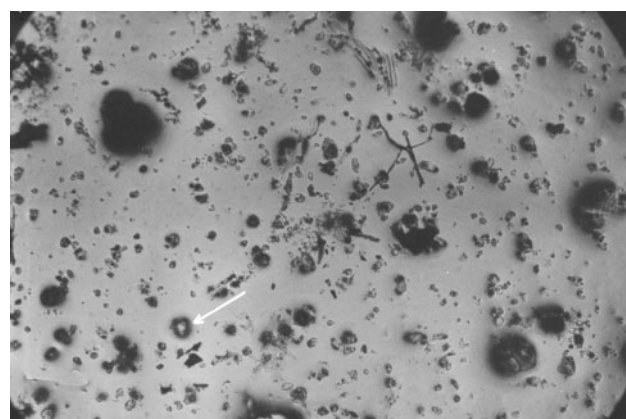


Fig. 2. Smoke particulates sampled with a rotary impactor (the arrow points to a particle of 10–15 μm in diameter).

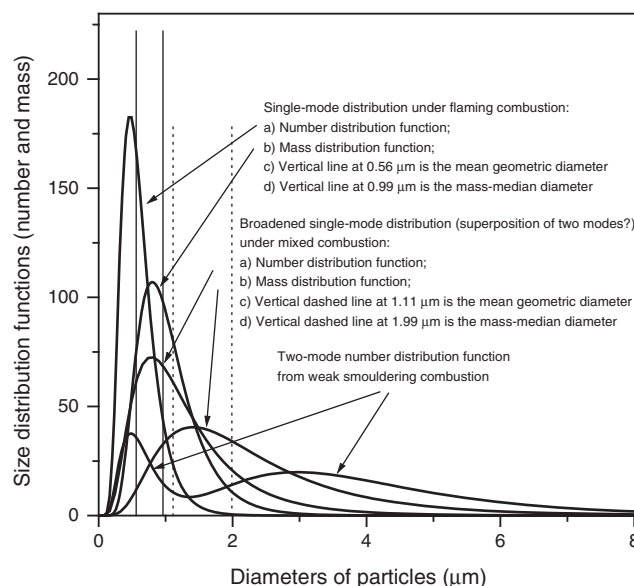


Fig. 3. Size distribution functions of fine fractions of smoke emissions under different combustion conditions. The number function is a percentage of particles per 1- μm size range, percentage of particles per micrometre. The mass function is a percentage of emission mass per 1- μm size range, percentage of mass per micrometre.

CaCO_3), are plate-like and very fragile. When a plate-like particle collides with the first impaction plate, the particle can split into smaller particles, which move forward and deposit on the next plates (this phenomenon is known; Fuchs 1964; Baron and Willeke 2001). Consequently, the true mass distribution, if sampling of coarse particulates by the cascade impactor was perfect, should contain somewhat more substance on the first, second and, to some degree, third plates (probably 5–10% in total instead of 2–5% in Fig. 1). Fig. 2 demonstrates the coarse particulates sampled with a rotary impactor, which collects just the coarse particles. The fire smoke is observed to contain many particles of 10–100 μm , which are sand (light objects), char and ash particulates (black objects).

The percentage of soil–mineral substances, $\sim 8\%$ on average, is in agreement with the revised total mass, 5–10%, deposited on the first, second and third impaction plates. The small particles of less than 3–5 μm (the fourth and fifth cascades) are dominant, making up 90–95% of the smoke matter. Measuring *in situ* the fine fractions of smoke with an optical particle counter–analyser demonstrates that they are mostly in the submicrometre and the 1- μm dispersal classes. Fig. 3 shows several examples of the number (N) distribution functions $\frac{100}{\sum N} \cdot \frac{\partial N}{\partial d}$ and the mass (M) distribution ones $\frac{100}{\sum M} \cdot \frac{\partial M}{\partial d}$, calculated from the counter data on the smoke generated under different combustion conditions. The distribution curves for fine emissions ranging from 0.2–0.3 to 5–8 μm in size (which correspond to the fifth and fourth impactor cascades in Fig. 1) indicate that the mean geometric sizes d_g were generally within 0.36–1.11 μm , but the mass-median sizes d_m were 1–2 μm . Under these conditions, the standard geometric deviation σ_g was ~ 1.6 – 1.7 for the log-normal size distributions in Fig. 3. We can estimate from these values that 92–98% of the smoke mass was in fine particles of less than 3–5 μm ($\leq d_m \sigma_g^2$), which agrees with the data in Fig. 1. These results are important for computational modelling of the influence of fire smoke on solar heat transfer in the atmosphere because smoke is found to be composed mainly of submicrometre and 1- μm particles, which strongly affect both sunlight scattering and the residence time of particles in the air. Similar size distributions of smoke particulates were found in fires in North America and Africa (Radke *et al.* 1988, 1991; Cachier *et al.* 1989, 1991).

Morphological properties and specificity of trace element composition of fine particulates

A common question is whether there is any correlation between the energy release (fire intensity) of a forest fire and the chemical and dispersal compositions of smoke particles. Such interrelations were often observed in test-bench experiments (Hao *et al.* 1991; Lobert and Warnatz 1993). In actual fires, the irregular changes in combustion conditions (flaming or smouldering) and, consequently, in smoke production, especially as related to the variations of wind power and direction, make the determination of this empirical linkage difficult. Our model fires indicate that the amount of smoke produced by flaming burning is visually less than that from smouldering combustion. In this respect, two successive fires in 2002 were indicative of this. One of the fires was of higher power (5000 kW m^{-1}), whereas the second fire was of moderate intensity (3000 kW m^{-1}). More than a two-fold decrease in mass concentration of smoke particles was observed in the high-intensity fire, and the smoke particles deposited on the filters were of visually different morphological structures. A plausible explanation for reduced smoke yield on flaming combustion is that both the tiny granules of burning vegetation lifted by the upward fire streams and the vapours of organic compounds (tars, lignin and others) cannot cross a wide and high zone of flaming combustion without being consumed. This should result in reduced amounts of unburned organic matter, i.e. in a decrease of smoke concentration.

In the model fires of 2008–10 performed on the $6 \times 12\text{-m}$ plots, we had an opportunity to choose sites with uniform distributions of fuels over the plots. Under these conditions, the burning and smoking processes proceeded more steadily

during the fire. We observed that the emissions sampled over the flaming zone differed visually from the particles produced by smouldering combustion. The filters with ‘flaming’ particles were of coal-black colour, and the mass of emissions sampled on the filters were substantially less than those from smouldering combustion. The ‘smouldering’ filters were of brown and yellow colours, and the particulate matter was oily (this is due to the incomplete burning of the tar and lignin vapours in the zone of smouldering combustion). A systematic relationship between fire conditions and particulate mass also follows from the data in Fig. 1. The percentage of the mass sampled on the fourth cascade (particles 3–7 μm) ranged from 1 to 13% ($6.9 \pm 5.9\%$). However, this wide dispersion is generally caused by combustion conditions rather than by trivial sampling and measuring inaccuracies. The emission pumped from a flaming zone gave only a 1–3% percentage, but the 8–13% share was in the case of smouldering combustion. In the latter case, the particles deposited on the fourth impaction cascade (a glass plate coated with sticky matter) looked like round and oily particles (for a photo of these particles, see Samsonov *et al.* 2010).

Table 3 presents the trace element composition of fine smoke particles sampled by the fifth impaction cascade (the polymeric AFACHA filters). These particles of ~ 0.1 – $3.0 \mu\text{m}$ in diameter formed from the unburned vapours of organic compounds (tars, lignin and thermally decomposed cellulose) that evaporated or sublimed from the heated biomass. The major portion of vapours (~ 93 – 99% as follows from a smoke factor of 1–7%) was consumed but a certain amount avoided combustion. The ingress of unburned vapours into the relatively cold air above the combustion zone was followed by nucleation, condensation and coagulation processes resulting in the above fine particles. On biomass burning, the potassium, zinc, calcium and other ‘vital’ elements contained in the biomass tissues were released into the air in the form of chemical salts and oxides. These substances could be then incorporated into the condensation and coagulation particles. Thus, the amounts of elements found in the fine particulates characterise both the initial proportions of elements in plant tissues and their ability to be incorporated into particles.

The trace element concentrations presented in Table 3 were averaged for several filters taken from the fifth impaction cascade but were then normalised to the mean potassium content. The mean potassium content was conditionally attributed to be 100 dimensionless units. Consequently, the relative concentrations of all elements in the table are dimensionless too. Table 3 shows also the normalised concentrations of the elements sampled by pumping smoke directly through the AFACHA filters (i.e. without the impaction facility). As indicated in this table, the normalised concentrations of some elements (V, Ti, Fe and others) on the fifth cascade are appreciably lower than those sampled using direct sampling (i.e. the ‘fifth cascade/direct sampling’ ratio is less than unity). This supports the hypothesis that these elements take little part in any biological functions. Consequently, biomass combustion produces not much of these elements in fine particles but mainly in coarse particulates originating from soil. Such elements as potassium, zinc, copper, bromine, arsenic and calcium to some extent are plant nutrients, so that the increased concentrations of these

elements in fine smoke are expected (the above ratio is equal to or greater than unity). The contents of manganese and molybdenum in fine aerosols were found to be low as those from the direct sampling. This is quite strange, as these elements are known to be the plant nutrients, and one might expect increased concentrations of these in fine particulates. In Samsonov *et al.* (2005), the enrichment factor was used to discern whether these elements had originated from vegetation tissues. The affirmative conclusion of this in the earlier paper is at variance with the low levels of molybdenum and manganese found in fine particles in the present work. Possibly, during biomass combustion, these elements (the chemical salts and oxides) have become concentrated mostly in the residual ash matter and become depleted the fine particulates. Gaudichet *et al.* (1995) and Echalar *et al.* (1995) reported that potassium and bromine are incorporated almost fully in fine submicrometre particles (our data agree with this conclusion). According to the cited data, more than 90% of zinc originates from forest soils, i.e. zinc is incorporated in the coarse particulates. Our data (Table 3), however, show that the burned biomass tissues produce ~60% of zinc in the form of fine particles. This indicates that the enrichment factor method fails sometimes, but the multi-elemental composition of fine particles seems to be more reliable in determining the origins of these elements in the air.

Conclusion

Study on the chemical composition, dispersal parameters and mass capacities of smoke emissions leads to the conclusion that there were no essential differences in the mass, chemical and dispersal characteristics of the particulates found in the smoke from fires in two forest types in central Siberia (pine and larch mixedwood). This is in agreement with our previous results and the reference data on fires in North America that the particulate smoke composition is strongly dependent on the burn efficiency of fuels (dryness factor) and the combustion conditions (flaming or smouldering combustion), and, to a lesser extent, the type of vegetation burned. However, the 0.1–3 and 3–7- μm smoke particles (sampled on the fifth and fourth impaction plates) manifest specific features of dispersal composition and morphological structure depending on the combustion properties (fire intensity, spatial length of combustion zone) and vegetation types. In particular, smouldering combustion of coniferous fuels produces copious amounts of submicrometre particles that contain oily substances. These particles can efficiently coagulate with each other, forming agglomerates of 1–5 μm in diameter, which can contain a considerable proportion of oily (tarry) matter. Intensive flaming combustion mainly forms dry (non-oily) particles of submicrometre classes. The elemental composition of fine fractions of smoke particulates can discriminate between the origins of different elements.

The data on the dispersal parameters of smoke particulates are important for the computational modelling of the influence of fire smoke on solar heat transfer in the atmosphere because smoke is found to be composed mainly of submicrometre and 1- μm particles, which strongly affect both sunlight scattering and the residence time of particles in the air. The amount of fine particles of less than 2.5 μm in size ($\text{PM}_{2.5}$) is commonly used to

characterise the respiratory quality of near-land air as such particles can easily penetrate through a human rhinopharynx into the lungs. One can see that such particles contribute ~90% of the total mass of smoke emissions (the inequality of the 2.5- and 3- μm operating cut-offs should be considered negligible taking into account the geometrically irregular shapes of smoke particulates). Besides, smoke particulates contain 8–10% of elemental carbon (this matter is also called soot and black carbon, which are not always the same), which is accepted as a dangerous atmospheric pollutant. In this regard, smoke emission from a large-scale forest fire can induce negative effects on human health over a wide area.

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