Particulate emissions from fires in central Siberian Scots pine forests[1]

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Abstract: Siberian boreal forest fires burn large areas annually, resulting in smoke that releases large amounts of particulate emission into the atmosphere. We sampled aerosol emissions from experimental fires on three Scots pine (*Pinus sylvestris* L.) forest sites of central Siberia. Emissions from ground-based aerosol samples were 0.1–0.7 t/ha. This value represented 1%–7% of the total biomass (10–30 t/ha) consumed during the experimental fires. We were able to classify the chemical composition of 77%–90% of the mass of particulate fire emissions. Chemical analysis indicated that an average of 8%–17% of the particulate composition was of mineral origin. Carbonaceous aerosols created because of incomplete combustion ranged from 50% to 70% of the total aerosol mass. The fraction of aerosols containing elemental carbon (EC) (i.e., graphite, soot, and charcoal) was 7%–15%. As our samples were taken near the ground surface, these results represent freshly emitted fire aerosols that have not yet had time to react with atmospheric moisture or to undergo postfire chemical or physical–chemical changes. In a typical year, where 12×10^6 – 14×10^6 ha burn in Russia, we estimate that 3×10^6 – 10×10^6 t of particulate matter may be emitted into the atmosphere.

Résumé : À chaque année, les incendies brûlent de vastes zones de forêt boréale en Sibérie. Ces incendies produisent de la fumée qui relâche dans l'atmosphère de grandes quantités de particules. Ils ont échantillonné les émissions d'aérosols provenant de feux expérimentaux dans trois forêts de pin sylvestre (*Pinus sylvestris* L.) du centre de la Sibérie. Sur la base d'échantillons d'aérosols prélevés au sol, les émissions atteignent 0,1–0,7 t/ha. Cette valeur représente 1 % – 7 % de la biomasse totale (10–30 t/ha) consumée par les feux expérimentaux. Les auteurs ont pu classer la composition chimique de 70 % – 90 % de la masse des émissions de particules dues au feu. L'analyse chimique a montré que 8 % – 17 % des particules sont d'origine minérale. Les aérosols carbonés provenant de la combustion incomplète comptent pour 50 % – 70 % de la masse totale d'aérosols. La proportion des aérosols contenant du carbone élémentaire (c.-à-d. du graphite, de la suie et du charbon) est de 7 % – 15 %. Étant donné que nos échantillons ont été prélevés près de la surface du sol, ces résultats représentent les aérosols fraîchement émis par le feu qui n'ont pas eu le temps de réagir avec l'humidité atmosphérique ou subir des modifications chimiques ou physico-chimiques à la suite du feu. Au cours d'une année typique en Russie, où 12×10^6 – 14×10^6 ha de forêt sont détruites par le feu, ils estiment que 3×10^6 – 10×10^6 t de particules seraient émises dans l'atmosphère.

[Traduit par la Rédaction]

Introduction

The burning of forest biomass (grass, moss, lichen, shrub, and wood) results in atmospheric emissions of large quantities of gases (e.g., CO_2, CO, CH_4, etc.) and smoke particles. Particulate emission is composed of aerosols of various sizes, from submicrometre to coarse particles tens of micrometres in diameter. These smoke particles of different sizes display a

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range of ecological, chemical, physical, and optical properties due to their different physical and chemical composition.

Convection of hot gases from forest fires, which entrain mineral and organic soil particles from the ground layer, contributes to aerosol emissions. Entrained aerosols include soil particles that have accumulated prior to the fire as dust on vegetation or on surfaces of litter and ground fuels. Because these soil and dust particles are subjected to strong heating in the fire zone, where most of their organic substrate material is consumed, the particles become almost fully mineralized. This appears to be the primary means by which fire emissions acquire a significant quantity of chemical elements typically found only in soils (e.g., iron (Fe), calcium (Ca), aluminum (Al), silicon (Si)). These soil-based elements in the particulate emissions are often referred to as either trace elements or microelements (Mason 1966; John et al. 1973; Rahn 1976; Kist 1987; Perelmann 1979). Some of these elements (e.g., potassium (K) and phosphorus (P)) are key nutrients for plant growth. During fires, these elements end up mostly in the ash layer on the soil surface (in the form of chemical salts and oxides), but some do escape as aerosols. A larger concentration of these elements than is characteristic of local soil concentrations may be found in the emissions. The soil and dust particles created from the burned ground layer and vegetation typically consist mainly of relatively coarse aerosols. Reported sizes range from 2 to 10 μm for clay particles and 10–60 μm for quartz ones (Morales 1979; Gillette et al. 1972; Ivlev 1991). Trace element emissions from the combustion of live and dead biomass tissue produces both coarse objects (such as ash flakes) and relatively small particles (<5–10 μm).

Carbonaceous aerosols produced from biomass burning normally range from submicrometre sized, formed in the course of the nucleation and condensation of carbon atoms and organic molecules and the coagulation of very small particles, to several micrometres, formed by the adsorption of carbon atoms and molecules on the surface of aerosols (Reid et al. 2005). Sizes can be tens of micrometres in diameter if the initial particles ejected into the fire zone were large (hundreds of micrometres). During burning, these particles decrease in size; nevertheless, their final sizes can still be relatively large (several or tens of micrometres). The production mechanism is similar for EC particles (i.e., carbonization of the initial biomass particles and condensation or adsorption of carbon atoms) with similar size ranges.

Fine (submicrometre-sized) aerosols, particularly those containing a large amount of EC, disturb normal radiation transfer in the atmosphere through their effects on sunlight extinction and cloud properties, which can affect local and global weather and climatic processes (Hobbs and Radke 1969; Reid et al. 1999). In this respect, the hypothetical climatic effect (atmospheric cooling) caused by the fire emission of carbon particles (Pittock et al. 1986) should compensate, to some extent, potential climatic warming due to increased CO_2 emissions. A proportion of fire emissions, consisting of both soil–mineral and carbonaceous particles, can also affect chemical processes occurring in the atmosphere through catalytic and photocatalytic reactions on the mineral particle surfaces (Parmon 1999). Such reactions can lead to smog formation; have effects on local, mesoscale, and global transportation of atmospheric chemical pollutants; and have effects on air quality (Crutzen and Andreae 1990; Houghton et al. 2001; Kondratyev and Isidorov 2001).

This paper reports on the mass concentrations of aerosols in smoke emissions of experimental fires in boreal Scots pine (*Pinus sylvestris* L.) forests of central Siberia. In addition, we report on the trace elements content of these particles, on the organic substances, EC, and mineral components found in the fire aerosol emissions, and on the emission particle sizes observed.

Methodology

Field experimental design

Smoke emission sampling was conducted as part of the Russian FIRE BEAR (Fire Effects in the Boreal Eurasia Region) Project, which studies forest fires of central Siberia. The project was developed to provide important answers to problems associated with the management of fuels, fire, and fire regimes, to enhance carbon storage and forest sustainability in ways that minimize the negative impacts of fire on the global environment, wood production, and ecosystem health (McRae et al. 2005). The research sites are located in the Krasnoyarsk region of central Siberia. All sites are named for nearby villages. The Yartsevo site is west of the Yenisey River (60°38′N, 89°41′E); the Govorkova and Khrebtova sites are, respectively, south of the Angara River (58°35′N, 98°55′E) and north of the Angara River (58°25′N, 98°25′E). Stands in these research areas are representative of central taiga pine forest (Parmuzin 1985). The particular forest type represented on our research sites is a Scots pine / lichen (*Cladonia* sp.) / feather moss (*Pleurozium schreberi* (Brid.) Mitt.) forest. The soils are sandy alluvial-ferrous Podzols supported by small-grained carbonate-free sand. These forests are site quality classes IV and V (Anuchin 1982), which indicate that forest productivity is poor. There is no underlying permafrost.

The experimental fire plots were 4 ha (200 m × 200 m) except at Govorkova, where the two plots were each 2.3 and 3.1 ha. Experimental fires were carried out in June and July of 2000–2003, which are the typical burning months for this region. Fire management personnel of the Russian Aerial Forest Protection Service (Avialesookhrana) and Khrebtova Forestry were responsible for the construction of the protective fire lines (consisting of plowlines) and the actual ignition and suppression of the fires. All experimental plots were burned using line ignition along the windward side to quickly create equilibrium fire behavior that mimicked wildfires under similar burning conditions (McRae 1996). Ignition commenced in the middle of the windward plot side and was carried out by two ignition teams using Russian driptorches walking quickly to either plot corner to ensure rapid ignition of the entire side. The driptorches were also used to burn out each side as the fire spread down the plot to ensure containment of fire within the fire lines. In more severe burning conditions, the fire lines were widened on critical sides with a 5-m burnout strip prior to ignition of the main fire. Before each fire a full inventory of preburn fuels was performed. During the fires, complete documentation of fire behavior (e.g., fuel consumption, rates of spread, fireline intensity, etc.) was carried out. Details on the inventory and documentation process are described in McRae et al. (2005).

Fig. 1. Fire aerosol samples being taken during the smoldering phases of combustion at ground level.

Emission sampling was carried out on every fire. Fires are identified according to the numbering scheme of the plots at each site (e.g., fire 5 occurred on plot 5). Plots were not burned in any numerical order. At Yartsevo, plots were burned in 2000, 2001, and 2002.

Field sampling of aerosols on filters

We sampled particulates from the ground at the outer edges of the experimental burning plots (Fig. 1). Sampling locations were periodically moved 20–25 m, as the research team followed the progression of the fire across the plot. Smoke was sampled through a circular 70-mm polymeric thin-fibrous aerosol filter (Petranov AFAChA 20-cm^2 filter) and a 50-mm glass-fiber filter (Gelman 15-cm^2 filter) that were held in a single holder 0.4 m apart. A Y-fitting attached each filter to a single pump with a 20 L/min flow rate. Collection times ranged from 2 to 10 min, with the filter inlet of the holder located at a height of 0.5–1.0 m over the smoke sources. AFAChA filters are used to measure the partial quantities of trace elements, while the Gelman filters are used to detect carbonaceous components. A record was kept of pertinent information for each filter sample (e.g., date of the sample, sample location, fire conditions, kind of fuel burned, pumping flow rate, and pumping time).

Aerosol sample analysis

Filters were dried (5–7 days) to a constant mass in a sealed glass dessicator filled with granules of NaX zeolite that had been dried at 400 °C for several hours. Dried filters were weighed prior to use in a closed Petri dish to avoid reabsorption of ambient moisture. After the fires, used filters were reweighed using these same procedures. Any filter mass increase was attributed to smoke particulate accumulation. The net mass of particulates on the filters ranged from 0.5 to 10 mg depending upon the smoke characteristics and aspiration time. Carbonaceous organic substances sampled on the glass filters (Gelman) were measured using reaction gas chromatography (Makarov et al. 1996). One-quarter to one-half of the glass filter was placed on a sampling dish, which was placed into an argon (Ar) flux at 700 °C for 20–30 s. At this temperature all organic substances on the filter sublimate and enter the catalytic reactor, where the organic compounds are oxidized into CO_2. The CO_2 is subsequently separated in a chromatographic column and passes through a multipass valve to the catalytic reactor to be converted into CH_4. The resulting CH_4 is measured quantitatively with a flame-ionization detector. The principle of this method is to successively convert each carbon atom from an organic molecule to CO_2 by oxidation, and then into CH_4 (i.e., a one-carbon molecule). Hence, this method measures the total mass of carbon atoms contained in the organic compounds collected on the samples regardless of their original chemical structures. The carbon mass in a sample is determined using stearic acid as a calibration compound. However, in calculating the total mass of organic substances in a sample one must take into account the fact that in addition to carbon atoms, those organic molecules can contain atoms of oxygen (O), hydrogen (H), and nitrogen (N). The organic components of smoke aerosols should consist of products created from cellulose, hemicellulose, lignins, and resins. The mass fraction of carbon atoms in these compounds ranges from 45% to 55%, with a 50% average (Konev 1977; Levine and Cofer 2000; Levine et al. 2000). Therefore, we doubled the measured carbon mass to estimate the total organic mass of an aerosol sample.

The basic chromatography apparatus used in determining organic carbon (OC) was also used to measure EC on the

Table 1. Total mass concentration (C_0) of aerosol emission and the partial concentrations of some trace elements from 2001 ground sampling of forest fire smoke taken at Yartseva.

AFAChA filter sample No.	C_0 (mg/m^3)	Trace element (µg/m^3)														
		K	Ca	Ti	Cr	Mn	Fe	Co	Zn	As	Se	Br	Rb	Sr	Mo	Pb
Yartsevo fire 2																
5	244.0	85	139	0.0	0.0	9.5	25.4	0.32	4.9	0.00	0.00	0.6	0.2	0.49	0.00	0.9
6	35.7	439	54	0.0	5.0	0.4	22.5	1.04	6.1	0.00	0.12	2.6	1.8	0.06	0.25	3.1
7	38.5	233	100	2.1	1.9	11.0	49.2	0.31	3.3	0.94	0.00	0.8	0.9	0.14	0.09	0.0
9	30.0	23	39	0.0	1.7	0.3	7.5	0.02	0.0	0.12	0.00	0.5	0.0	0.05	0.05	0.2
10	78.1	144	53	0.0	5.6	3.8	33.1	0.00	0.0	0.00	0.00	0.4	0.0	0.13	0.38	1.0
Fire mean	85.3	185	77	0.4	2.8	5.0	27.5	0.34	2.9	0.21	0.02	1.0	0.6	0.17	0.15	1.0
Yartsevo fire 3																
17	37.7	455	38	0.0	0.0	2.3	10.0	0.00	14.5	0.23	0.26	2.3	1.8	0.00	0.00	1.3
20	22.0	88	70	1.8	2.6	19.6	9.5	0.11	0.0	0.00	0.00	0.2	0.3	0.10	0.00	1.0
21	37.0	106	25	0.0	0.0	1.2	17.5	0.00	0.7	0.00	0.00	0.6	0.4	0.00	0.00	0.0
23	63.8	25	102	14.5	11.9	8.5	24.7	0.00	0.0	0.00	0.00	0.0	0.0	0.00	0.17	0.6
25	42.1	185	61	3.0	0.0	4.6	11.4	0.00	1.7	0.27	0.13	0.9	0.8	0.03	0.12	0.7
27	56.6	25	34	0.0	0.0	0.0	16.6	0.00	0.8	0.10	0.00	0.2	0.2	0.00	0.11	0.9
28	29.1	206	274	16.5	5.2	10.9	13.4	0.13	2.7	0.80	0.00	0.4	0.6	0.20	0.00	0.0
29	76.2	30	53	6.5	0.0	6.1	12.4	0.00	1.2	0.70	0.06	0.5	0.1	0.42	0.00	0.1
Fire mean	45.6	140	82	5.4	2.5	6.7	14.0	0.00	2.7	0.20	0.00	0.6	0.5	0.00	0.00	0.6
Yartsevo fire 6																
16	76.9	915	46	0.0	7.7	0.0	69.2	1.77	2.3	0.85	0.24	5.8	4.8	0.09	0.85	3.7
32	50.0	205	65	0.0	8.5	0.0	5.0	0.23	1.5	0.35	0.25	1.1	0.8	0.00	0.27	0.9
34	31.9	364	311	12.8	5.3	25.7	19.6	0.00	6.2	0.00	0.00	2.3	1.1	0.61	0.00	0.9
45	32.8	398	39	0.0	0.0	1.5	9.2	0.00	2.6	0.00	0.00	0.4	1.6	0.00	0.00	0.0
46	30.0	619	49	1.8	0.0	9.0	4.1	0.24	5.7	0.00	0.00	1.9	2.8	0.11	0.13	0.8
47	20.8	54	94	0.0	1.9	5.6	7.1	0.08	0.0	0.00	0.13	0.2	0.2	0.15	0.00	0.6
48	15.4	54	77	0.0	0.0	8.9	12.0	0.00	0.6	0.00	0.00	0.0	0.1	0.17	0.00	0.0
Fire mean	36.8	373	97	2.1	3.3	7.2	18.0	0.33	2.7	0.17	0.09	1.7	1.6	0.16	0.18	1.0
Yarstevo fire 19																
51	48.2	22	29	1.7	2.2	0.0	6.0	0.27	0.0	0.00	0.17	0.1	0.2	0.00	0.00	0.7
52	111.1	83	10	2.9	0.0	1.0	15.6	0.00	0.0	0.00	0.00	0.3	0.5	0.18	0.00	1.5
56	74.4	815	142	8.9	3.5	12.2	13.4	0.13	6.9	0.23	0.10	2.4	3.0	0.20	0.15	0.9
57	40.0	685	125	0.0	0.0	12.0	15.6	0.37	5.3	0.15	0.05	3.6	3.5	0.20	0.04	1.4
58	29.8	412	17	3.9	0.0	1.8	8.2	0.33	5.3	0.26	0.12	3.9	1.7	0.00	0.00	2.6
Fire mean	60.7	403	64.6	3.5	1.1	5.4	11.8	0.22	3.5	0.13	0.09	2.1	1.8	0.12	0.04	1.4
2001 total mean	54.1	267	82	3.1	2.5	6.2	17.5	0.21	2.9	0.20	0.07	1.3	1.1	0.13	0.10	1.0
Standard error	9.1	53	15	1.0	0.7	1.3	2.9	0.08	0.7	0.06	0.02	0.3	0.3	0.03	0.04	0.2

same piece of filter sample. An Ar–O_2 flux was used for this test. The O_2 reacts with the EC to form CO_2 at 700 °C. The resulting Ar–O_2–CO_2 mixture passes through a chromatographic column, where the CO_2 enters a catalytic reactor (methanator) to be converted into CH_4 and is measured using a flame-ionization detector. In this case pure graphite was the calibration compound used to determine EC mass. This indirect method was used, since temperatures exceeding 3000 °C would be required to vaporize EC in the absence of O_2.

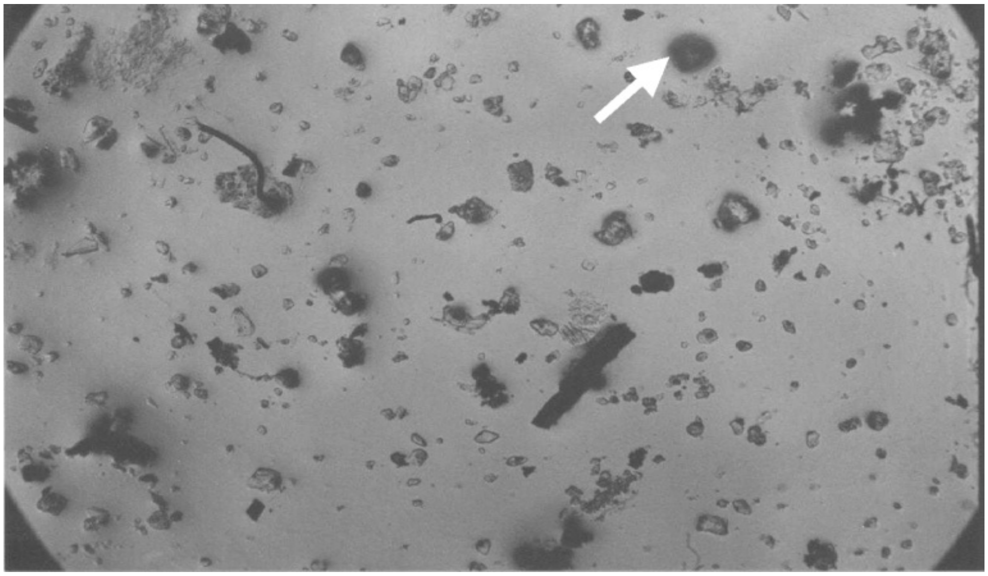
Trace elements of the aerosols collected on the AFAChA filters were determined by the synchrotron radiation X-ray fluorescence (SRXRF) method (Barashev et al. 1995). This method can detect elements with atomic masses only heavier than K. Detection limits ranged from 0.05 to 0.1 µg/cm^2 for Ca and K and 0.0004–0.001 µg/cm^2 for strontium (Sr), zirconium (Zr), and molybdenum (Mo). If the SRXRF method resulted in a concentration for a specific element less than the detection limits, a zero value was nominally assigned, even though a trace of it might exist in the sample (Tables 1 and 2).

We determined the trace elements derived primarily from the burning of biomass tissues versus those primarily produced from mineral particles derived from the soil using the "enrichment factor" procedure of Rahn (1976) and Mason (1966). The enrichment factor of an element is the ratio of its normalized concentration in an aerosol sample to the normalized concentration found in the soil. The normalized concentration is the ratio of the element concentration to the concentration of Fe or Al (whose presence would be solely due to soil particle emissions). A comparison of the normalized concentrations found in aerosol emissions and natural soils allows us to calculate the "aerosol/soil" ratio for any given element, which is called the enrichment factor. If this

Table 2. Total mass concentration (C_0) of aerosol emission and the partial concentrations of some trace elements from 2003 ground sampling of forest fire smoke taken at Khrebtova.

AFAChA filter sample No.	C_0 (mg/m^3)	Trace element (μg/m^3) K	Ca	Ti	Cr	Mn	Fe	Co	Ni	Cu	Zn	Br	Rb	Sr	Mo	Pb
Yartsevo fire 3																
6	40.0	188	781	10	1.1	57	41	0.3	1.0	1.8	4.4	0.3	0.1	1.7	0.0	0.3
7	68.0	275	577	72	1.9	41	274	1.3	1.8	7.3	13.8	1.4	0.5	2.2	0.0	0.8
8	28.0	282	424	119	4.6	42	366	2.1	3.5	5.9	6.2	1.8	0.5	2.0	0.0	1.5
Fire mean	45.3	248	594	67	2.5	47	227	1.2	2.1	5.0	8.1	1.2	0.37	2.0	0.0	0.9
Yartsevo fire 5																
2	95.0	759	1809	76	0.7	163	258	1.0	1.1	2.6	7.1	0.7	0.7	3.8	0.1	0.7
3	29.0	309	775	25	1.0	78	88	0.5	0.9	1.2	3.4	0.3	0.2	1.2	0.0	0.5
4	46.0	454	1362	62	0.7	123	193	0.6	1.0	1.0	4.1	0.3	0.4	2.6	0.1	0.4
5	56.0	110	216	15	1.2	10	59	0.4	1.2	2.3	2.9	0.5	0.1	0.6	0.1	0.3
Fire mean	56.5	408	1041	45	0.9	94	150	0.6	1.1	1.8	4.4	0.5	0.4	2.1	0.1	0.5
2003 total mean	51.7	339	849	54	1.6	73	183	0.9	1.5	3.2	6.0	0.8	0.4	2.0	0.04	0.6
Standard error	9.0	81	210	15	0.5	20	47	0.2	0.4	0.9	1.4	0.2	0.1	0.4	0.02	0.2

Fig. 2. A close-up of 2002 smoke particles collected on an impactor glass plate at Yarstevo. The white arrow points to a round dark particle at the top of the photograph with a diameter of about 15–20 μm.



factor exceeds 10, natural soil is not considered a source of the element. In fire emissions, elements with large enrichment factors can only originate from burned plant tissues, which contain K, zinc (Zn), and other vital elements. If the enrichment factor is not large (e.g., <2), soil is the probable source of these elements. Reference data on the normalized concentrations for typical natural soils (clay, sand, rock material, sedimentary material, shale) were taken from Kist (1987) and Perelmann (1979).

Aerosol sampling by inertial impactor

For the 2002 and 2003 fire experiments, aerosol emissions were sampled on glass plates of a rotary impactor. The impactor design consists of an electric motor that rotates an Al rod with shoulders 100-mm long at a rate of about 1500 cycles/min. Two glass plates (5 mm × 25 mm) are fixed at the ends of each shoulder. We coated the impaction plates with a thin layer of viscous liquid (Apiezon grease) to provide a sticky surface for smoke particles to adhere to. The rotating impactor was positioned in the smoke for 0.5–1.0 min for each sample taken. The deposited particles were photographed in the laboratory. Digital images of ensembles of particles from impactor plates (Fig. 2) were analyzed using GIS to determine size parameters of individual particles (diameter or width, perimeter, projected area) and to calculate descriptive statistics for the whole ensemble (i.e., the mean size and the standard deviation). The "open" inertial impactor we used has the advantage, compared with "closed" type impactors, of efficiently capturing relatively coarse aerosols (e.g., of 30 μm and more) with minimal loss. On the other hand, only particles with diameters greater than 5–7 μm are trapped effectively (≥50%) on the plates of our device. Smaller particles

Fig. 3. A view of a typical surface fire (Govorkova fire 1) on a central Siberian Scots pine site.

(4–5 µm in diameter) are captured with less than 50% efficiency, while sampling of particle diameters <3 µm is not practical.

Results and discussion

Fires

All experimental burns sampled were surface fires of varying intensity and severity (Fig. 3). Since roughly 80% of Siberian fires burn as surface fires during typical burning conditions (Belov 1976; Furyaev 1996; Korovin 1996), our aerosol estimates should be considered reasonable for most fires on Scots pine sites across Siberia. McRae et al. (2005) report details of the fire behavior characteristics of several of these experimental fires.

Total aerosol mass

Total mass of the particulate emissions found on the AFAChA filters collected during the 2001 and 2003 fire experiments is shown in Tables 1 and 2. The aerosol total mass concentrations ranged from approximately 15 to 244 mg/m^3 (Table 1). Variations in mass concentrations resulted from nonuniform biomass burning (flaming versus smoldering combustion, fire duration and intensity) over the fire site. Variations reflected different surface fuels and combustion conditions (e.g., fuel moisture, wind speed and direction) both between burned plots and within plots. Such variations are typical for fires even on homogeneous sites. The sample location relative to the smoke source and smoke dispersal direction also affected this variability. Fluctuations in combustion (i.e., intensity of smoke formation) and variations in smoke drift direction over the 2- to 10-min sampling period for individual filters resulted in high variability in concentration of smoke at the sampling points. Even though the filter intakes were only 0.4 m apart, smoke quantities collected at the same time by the two filters could be quite different. This wide variation in particulate matter reflects the typical spatial and temporal variation in biomass smoke formation and wind (smoke) direction found under ambient (forest) conditions. By averaging the data collected during a single fire or for a set of fires for a particular year, we feel we gain a better estimate of characteristic emission concentrations for actual forest fire smoke production. However, individual samples are important in showing the spatial and temporal nonuniformity of biomass emissions.

Ground-level particulate total mass concentrations measured by the AFAChA filters averaged 54 and 52 mg/m^3 for the 2001 and 2003 fires, respectively (Tables 1 and 2). Although not shown, average total mass concentrations were 70 and 37 mg/m^3 for the 2000 and 2002 years, respectively. These values characterize the range in total mass concentrations of emissions produced by surface fires on our experimental sites and provide a basis for estimating the total particulate emissions entering the atmosphere during a typical surface fire in a Scots pine forest. Strong smoke formation occurred over a 5- to 30-min time period (τ) during both the flaming and smoldering combustion phases of our fires. Smoke generation peaked and then substantially decreased during this time period. During the smoke production period (τ), the observed velocity (U) of rising smoke (at a height of 0.5–1 m above the ground) was visually estimated to range from 0.5 to 1.0 m/s. Using a mean total mass concentration (C_0) of 50 mg/m^3 and an average velocity of 0.75 m/s, we can estimate that the total mass of emitted aerosols was $C_0U\tau \approx$ 11–68 g/m^2 (≈0.1–0.7 t/ha). Taking into account that the amount of actual biomass consumed during our experimental fires ranged from 10 to 30 t/ha (McRae at al. 2005), we estimate that during a typical surface fire on a Scots pine site 1%–7% of the consumed biomass is converted into particulate emissions.

Trace elements in smoke

The SRXRF method detected 15–20 trace elements in the smoke emissions (Tables 1 and 2) ranging from K to lead (Pb). The partial concentrations of different elements in the fire emission varied over two or three orders of magnitude, which reflects the diverse natural concentrations of these elements in both local soils and biomass tissues. Trace element compositions of pre- and post-fire forest soils and plant tissues were reported previously (Chankina et al. 2001; Koutsenogii et al. 2002, 2003). We found that elemental

Table 3. Normalized aerosol concentrations (normalized to concentration of Fe) and enrichment factors (aerosol/soil ratio) of elements found for the 2001 Yarstevo fires.

	Trace elements													
Data	K	Ca	Ti	Cr	Mn	Fe	Co	Zn	As	Se	Br	Rb	Sr	Mo
Fire aerosol	14.6	4.6	0.25	0.13	0.32	1.0	0.01	0.18	0.0087	0.0033	0.13	0.06	0.007	0.005
Reference soil data	0.58	0.55	0.12	0.0023	0.02	1.0	0.0005	0.002	0.00012	0.00002	0.0007	0.004	0.001	0.0001
Enrichment factor	25	8	2	56	19	1	20	90	73	165	186	15	7	50

composition of fire aerosols was not directly correlated with data on either soil or plant elements. This fact is not surprising given that fire emissions do not have the same chemical and dispersal characteristics of soils, as given by Kist (1987), Perelmann (1979), Ivlev (1991), and Mason (1966). As a rule, clay soils consist of small particles (<10 µm in diameter) consisting mostly of Al_2O_3, SiO_2, and Fe_2O_3. In contrast, individual quartz particles (i.e., SiO_2 in sand) are usually large (10–60 µm). The upper forest soil layers, intermixed with living plant roots, dead plant debris, and other organic substances (humus), results in this layer being quite porous. During forest fires, this biomass can be consumed, leaving behind only the trace elements that originally composed it. Clay soils can disintegrate to particles <10 µm in diameter with the high fire temperature, but quartz particles can endure the heating without disintegrating. Consequently, the heating of the soil layer changes particle sizes depending upon its mineral constituents. As the ground fuels burn, the fire produces an upward gas flow through this layer, entraining clay, quartz, and other soil particles with it. The entrainment efficiency of the gas depends upon the size, chemical composition, and dispersal characteristics of the individual particles. Our sampling procedure does not allow us to understand the specific sources for the elements found in our emissions (i.e., whether elements originated from the soils, ash, or plant tissues).

We calculated enrichment factors for the mineral elements detected in samples from the 2001 Yartsevo fires (Table 1), and the normalized concentrations and enrichment factors are presented in Table 3. We concluded that K, Zn, chromium (Cr), selenium (Se), bromine (Br), Mo, and probably manganese (Mn) are derived from combustion of plant tissues. The enrichment factors for Cr, Zn, Se, and Br are quite high. The fuel components that burn, consisting of leaves, moss, lichens, needles, and litter, have high concentrations of nutrients compared with soil levels (Allen 1989). In the case of Br, it is not known whether high levels of particulate Br indicate high levels of emissions such as methyl bromide. The high enrichment factor of Se and Cr is more difficult to explain, as these are not plant nutrients. These metals (Se and Cr) do not have low boiling points. Their higher enrichment factors may indicate selective uptake by the native vegetation. At the same time, Fe and titanium (Ti) are likely to have originated from the soil. Ca and Sr may come from both sources, since their enrichment factors are in the 7–8 range.

The measured trace elements made up 0.5%–1.0% of the total mass of the aerosol emissions (Table 1). The SRXRF method detects only those elements with atomic masses (numbers) heavier than that of K (Tables 1 and 2). However, all soils include some lighter elements as either chemical salts or oxides (i.e., they combine with other elements such as O, N, sulphur (S), P, and inorganic carbon such as the CO_3 group. In addition, most soils include a large amount of salts and oxides of Al, sodium (Na), and Si that would be found in sand as SiO_2. Kist (1987) and Perelmann (1979) report that these “invisibles” of the SRXRF method are contained in most soils in larger amounts (10–20 times) than the elements summarized in Table 1. On this basis, we estimate that the total mass fraction of mineral components could be 5%–10% of the total mass of the aerosol emissions for our 2001 and 2002 fires, where the ground cover was dominated by lichen. For our 2003 fires (Table 2), the measured elements (K, Ca, Fe, Mn, Ti, and Zn) amounted to 2%–3% of the total particulate emission mass. Consequently, the total mass of substances of mineral origin may be 20%–40%. The main reason for this difference is that the Khrebtova site, where the 2003 fires occurred, had a living ground cover dominated by feather moss (*Pleurozium schreberi*) that was probably more dust laden (an additional argument for explaining this extra concentration of mineral and soil material will be discussed in the section on particulate size characteristics).

Carbonaceous substances in fire particulate emissions

Tables 4 and 5 summarize the aerosol concentrations of carbonaceous substances sampled on the Gelman glass-filters both in the form of organic chemical compounds (probably the products of thermochemical destruction of cellulose, resins, oils, and lignins) and EC (soot, graphite, charcoal). Results for all 2001 and 2002 fires are given in Tables 4 and 5, respectively. Ground-level particulate total mass concentrations measured by the Gelman filters averaged 53 and 29 mg/m^3 for the 2001 and 2002 fires, respectively (Tables 4 and 5). Although not shown, average total mass concentrations were 59 and 37 mg/m^3 for the 2000 and 2003 years, respectively.

The mean partial fraction of organic carbon (OC) ranged from 50% to 70% of the total aerosol mass. The mean fraction of EC in these same samples was 7%–15%. The average EC/OC ratio for the results in Table 4 was 0.17 (=12.4%/70.1%). Other results summarized for boreal forest studies (Reid et al. 2005) reported EC/OC ratios of 0.08, and 0.07 for flaming combustion. That our results were much higher may be due to analysis error for either the organic or inorganic component. The total carbon (Table 4) was greater than 100% for a number of samples, indicating a considerable error in some of the analyses. Taking into account the trace element component (5%–10% usually), we were able to classify the chemical composition of 70%–90% of particulate emissions in the smoke from our experimental burns. Because aerosols were sampled over actively burning areas at a height of about 0.5–1.0 m above ground, these results represent the chemical composition of aerosol emissions before any major reactions with atmospheric reagents have taken place. These

Table 4. Ground-level aerosol carbonaceous substance concentrations found in the 2001 particulate emissions at Yartsevo.

			Measured organic substance		Measured elemental carbon		
Gelman filter sample No.	Total mass concentration of aerosols (mg/m^3)	Total sample mass (mg)	Mass (mg)	Proportion (%)	Mass (mg)	Proportion (%)	Total fraction of carbonaceous substance (%)
Yartsevo fire 2							
5	109.1	8.9	7.0	79	1.36	15	94
7	20.6	2.1	2.5	123	0.62	30	153
9	40.5	7.9	5.0	63	0.67	8	72
10	46.3	3.0	3.6	123	0.53	18	140
Fire mean	54.1	5.48	4.5	97	0.80	17.8	115
Yartsevo fire 3							
17	48.1	5.1	2.6	51	0.69	13	65
21	13.4	1.1	1.1	102	0.10	4	106
23	48.5	1.9	2.6	135	0.44	23	158
25	22.3	2.1	2.1	99	0.19	9	107
27	56.7	3.1	2.3	76	0.55	18	94
28	20.2	2.0	1.5	76	0.39	19	95
29	81.8	8.2	4.0	49	1.75	21	70
32	98.2	3.9	0.7	17	0.21	5	22
34	41.5	3.9	1.5	37	0.44	11	48
Fire mean	47.9	3.5	2.0	71.3	0.52	13.7	85
Yartsevo fire 6							
45	50.3	6.0	3.0	49	0.79	13	62
46	35.5	7.1	3.9	55	1.50	21	76
47	52.1	5.0	2.6	52	0.69	14	65
48	37.6	4.9	4.1	83	0.31	6	89
56	82.1	9.9	5.8	58	1.67	17	75
Fire mean	51.5	6.6	3.9	59.4	0.99	14.2	73
Yartsevo fire 19							
51	24.1	2.0	0.7	36	0.10	3	40
52	140.9	8.9	4.9	56	1.06	12	67
53	38.2	6.0	3.5	59	0.43	7	66
54	65.2	9.8	6.4	65	1.17	12	77
57	52.2	7.9	3.9	49	1.06	13	63
58	35.5	6.0	3.6	61	0.85	14	75
Fire mean	59.4	6.8	3.8	54.3	0.77	10.2	65
2001 total mean	52.5	5.3	3.3	68.9	0.73	13.5	82.5
Standard error	6.3	0.6	0.4	6.0	0.10	1.3	6.7

data on the initial composition of fire particulate emissions are important as baseline data for understanding postfire physical–chemical transformations and their ecological consequences.

Mass imbalance (unclassified aerosol mass), which is the difference between the total sample mass and the sum of measured masses of organic, EC, and mineral emissions, was 10%–30% of the total aerosol emission mass. The presence of this imbalance indicated that we had underestimated the amount of SiO_2 (in the form of sand particles) in the fire emission. Additional measurements of SiO_2, carried out using atomic emission analysis on several aerosol samples (at the Institute of Inorganic Chemistry, Novosibirsk, Russia), confirmed that the samples actually contained an additional 3%–7% of mass as SiO_2. Adding this to the earlier 5%–10% value we used for estimating the fraction of mineral substances found in the emissions suggests that a more realistic estimate of the mineral and soil substances should be 8%–17% of sample mass. Even with this correction, we still had an imbalance of about 7%–23%. This discrepancy is likely due to a systematic inaccuracy in the measurement of OC by our method of thermal volatization and catalytic oxidation of organic compounds into CO_2. The problem with this method is the calibration uses only stearic acid, whereas organic substances can be quite different based on the thermal decomposition of cellulose, lignins, oils, and resins. The use of this single calibration compound may have led to systematic underestimates of organic substances in emission samples by as much as 10%–20%. Identifying an ideal calibration compound for this complex mixture of organic compounds of biomass remains a difficult challenge for future research.

Particulate size characteristics

The emission particulates from the 2002 fires illustrated in Fig. 3 are representative of the coarse-dispersal aerosol frac-

Table 5. Ground-level aerosol carbonaceous substance concentrations found in the 2002 particulate emissions at Govorkova and Yartsevo.

			Measured organic substance		Measured elemental carbon		
Gelman filter sample No.	Total mass concentration of aerosols (mg/m^3)	Total sample mass (mg)	Mass (mg)	Proportion (%)	Mass (mg)	Proportion (%)	Total fraction of carbonaceous substance (%)
Govorkova fire 1							
61	69.9	5.8	1.88	32	0.74	13	45
62	21.0	3.4	0.97	29	0.28	8	37
81	60.3	3.8	1.4	37	0.18	5	42
82	79.1	6.8	1.6	24	0.24	4	28
93	58.3	2.1	0.53	25	0.32	15	40
94	11.8	1.2	0.39	33	0.08	7	—
Fire mean	50.1	3.9	1.12	30	0.31	9	39
Govorkova fire 2							
63	12.7	1.8	0.53	29	0.08	4	33
64	43.9	3.6	1.83	51	0.36	10	61
65	16.4	2.4	0.64	27	0.09	4	31
66	24.5	2.6	0.57	22	0.08	3	25
67	11.0	1.7	0.52	31	0.06	4	34
68	8.0	1.6	0.74	46	0.08	5	51
Fire mean	19.4	2.3	0.81	34	0.13	5	39
Yartsevo fire 20							
1	17.1	2.4	1.15	48	0.25	11	59
3	8.4	1.3	0.36	27	0.04	3	30
5	10.9	1.8	1.44	80	0.24	13	93
6	14.7	2.2	1.26	57	0.12	5	62
Fire mean	12.8	19.3	1.05	53	0.16	8	61
Yartsevo fire 21							
2	18.4	1.0	0.23	23	0.09	9	32
4	25.7	1.7	0.84	50	0.05	3	53
7	54.3	1.9	1.63	86	0.15	8	94
8	22.0	1.8	0.69	38	0.16	9	47
Fire mean	30.1	1.6	0.84	49	0.11	7	56
Yartsevo fire 4							
9	80.9	3.8	1.95	51	0.36	10	61
10	16.2	1.2	0.72	60	0.05	4	64
11	24.6	1.5	0.82	54	0.23	15	69
12	6.4	0.7	0.57	81	0.05	7	88
17	13.9	1.0	0.75	73	0.06	6	79
Fire mean	28.4	1.6	0.96	64	0.07	7	72
2002 total mean	29.2	2.3	0.96	45	0.18	7.4	52
Standard error	4.7	0.3	0.10	4	0.03	0.8	4

tion, while particle sizes <3–5 μm were almost absent on the rotary impactor plates. The bulk of particles were in the range of 10–20 μm, along with some very coarse objects of about 50–100 μm. The mean particle size was between 15 and 20 μm. For the 2003 fires we estimated a much larger mean particle size of 50–60 μm. This difference was due to the larger amount of mineral and soil substance (20%–40%) in the 2003 fire emissions at Krebtova, as compared with the 2002 fires (5%–10%). The 2003 impaction images indicated that there were a larger number of dust particles in the smoke as a result of burning of the dusty moss ground cover. Dust particles (especially those from quartz sands) typically have a mean size of 50–60 μm (Gillette et al. 1972; Morales 1979; Ivlev 1991).

Particulate dispersal and the environment

Convecting gases of a forest fire easily uplift emission particles up to 40–100 μm in diameter. These particles reached a height of at least 1–2 km above ground from our small fires (based on unpublished helicopter data). On large fires in Scots pine, smoke can easily reach much higher altitudes (FIRESCAN Science Team 1996). Away from the influence of the fire, these particles will fall back to Earth (Stokes fall velocity is approximately 0.05–0.30, 0.003–0.012, and 0.0003 m/s for particles 40–100, 10–20, and 3 μm in diameter, respectively). Based on a plume height of 1–2 km and an upper wind velocity over the forest of 5 m/s (conservative estimates), particles <3 μm in diameter would remain in the air for many days and might be transported thousands of kilometres by the wind. Even particles 10–20 μm in diameter could be transported long distances. Heavier aerosols (e.g., 40–100 μm in diameter) would be deposited back to the ground within 30–300 km of the fire site. Consequently, fine aerosols up to 10 μm in diameter participate in global atmospheric transfer, whereas coarse fire emission particles are more important regionally (mesoscale) and locally. Each emission particulate size class and the distance of its transport could lead to effects on specific meteorological, physical–chemical, and (or) ecological processes. Regional influences of 10–30 μm diameter aerosols can result in weather changes (e.g., changes in fog, smog, rain, etc.), in catalytic reactions of chemical pollutants on the particle surface, and in effects on air quality (Seinfeld and Pandis 1997; Parmon 1999).

Conclusions

Carbonaceous aerosols, resulting from thermal decomposition of biomass accompanied with incomplete burning of their organic compounds, range from 50% to 70% of the total aerosol mass. The mean fraction of aerosols containing EC (i.e., graphite, soot, charcoal) is 7%–15%. The X-ray fluorescence and atomic emission analysis of the fire particulates collected on the filters indicates that an average of 8%–17% are of mineral and soil origin. Our results represent the chemical composition of aerosol emissions before any major reactions with the atmosphere have taken place. The aerosols sampled in this study had not yet had time to react with moisture and had not been subjected to any postfire chemical or physical–chemical conversions or reactions with pollutants or other compounds in the atmosphere.

We were able to determine chemical composition of 77%–90% of the particulate aerosol mass found in fire emissions. An additional refinement of the methodology will be necessary to characterize the remaining components. We estimated aerosol emission rates from surface fires in Scots pine at 100–700 kg/ha based on our experimental data, where emission concentrations ranged from 30 to 70 mg/m^3. This represented about 1%–7% of the total mass of the consumed biomass (10–30 t/ha) during our experimental fires, which represented a range of burning conditions typical of forest wildfires on pine sites in boreal Russia. If we extrapolate these results more broadly to the $12 \times 10^6 - 14 \times 10^6$ ha burned each year by fires in Russia (Cahoon et al. 1994; Conard and Ivanova 1997) and assume that emissions are similar for other types of vegetation, we can estimate that $3 \times 10^6 - 10 \times 10^6$ t of particulate aerosols are emitted into the atmosphere annually from wildfires in Russia under typical burning conditions. Much of this material can be expected to enter into the general atmospheric circulation, affecting global energy balance, regional haze, and local air quality. Additional research is necessary to accurately estimate aerosol emissions from crown fire and for surface fires in other vegetation types and regions of Russia.

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