AGU PUBLICATIONS

JGR

Journal of Geophysical Research: Atmospheres

RESEARCH ARTICLE
10.1002/2017JD026825

Special Section:
Quantifying the emission, properties, and diverse impacts of wildfire smoke

Distribution of Trace Gases and Aerosols in the Troposphere Over Siberia During Wildfires of Summer 2012

P. N. Antokhin[1], V. G. Arshinova[1], M. Y. Arshinov[1], B. D. Belan[1], S. B. Belan[1], D. K. Davydov[1], G. A. Ivlev[1], A. V. Fofonov[1], A. V. Kozlov[1], J.-D. Paris[2], P. Nedelec[3], T. M. Rasskazchikova[1], D. E. Savkin[1], D. V. Simonenkov[1], T. K. Sklyadneva[1], and G. N. Tolmachev[1]

[1]V.E. Zuev Institute of Atmospheric Optics SB RAS, Tomsk, Russia, [2]Laboratoire des Sciences du Climat et de l'Environnement/IPSL, CNRS-CEA-UVSQ, Gif-sur-Yvette, France, [3]Laboratoire d'Aerologie, CNRS-UPS, Toulouse, France

Key Points:
- Emissions from Siberian biomass burning can significantly affect polar region environments and enhance radiative forcing
- Comprehensive in situ measurements of the vertical distribution of atmospheric constituents during the events of extensive wildfires in Siberia are still lacking

Correspondence to:
B. D. Belan,
bbd@iao.ru

Citation:
Antokhin, P. N., Arshinova, V. G., Arshinov, M. Y., Belan, B. D., Belan, S. B., Davydov, D. K., et al. (2018). Distribution of trace gases and aerosols in the troposphere over Siberia during wildfires of summer 2012. *Journal of Geophysical Research: Atmospheres*, *123*, 2285–2297. https://doi.org/10.1002/2017JD026825

Received 22 MAR 2017
Accepted 31 JAN 2018
Accepted article online 7 FEB 2018
Published online 20 FEB 2018

Abstract The results of sensing of the gas and aerosol composition of the atmosphere with the Optik Tu-134 aircraft laboratory in the period from 31 July to 1 August 2012 are presented. The measurements were conducted along the flight route Novosibirsk-Tomsk-Mirny-Yakutsk-Bratsk-Novosibirsk. A significant part of the Siberian territory during this period was covered by numerous forest fires. The synoptic situation during the measurements was characterized by the presence of low-gradient field. This fact determined the low rate of transport and diffusion of pollutants and their accumulation in the region under study. The maximal concentrations of CO_2, CH_4, and CO over fire zones achieved 432 ppm, 2367 ppb, and 4036 ppb, respectively. The aerosol particle number density in emission plumes achieved 4400 cm^{-3}. Outside emission plumes, the concentration ranged within 400–1000 cm^{-3} depending on the region. The mass concentration of aerosol in plumes increased approximately 7 times (6.9). The enrichment of the concentration of some elements and ions in the plume with respect to the background varied from 1.3 to 9.1 times. The generation of ozone from biomass burning products was observed at plume boundaries. Two versions of this process are possible: ozone generation under and above the plume.

1. Introduction

Historically, first traces of forest fires on the Earth were discovered in paleoclimatic data and are dated to the period of 350–400 million years ago (Andreae, 1991). In the process of biomass burning during forest fires, huge amounts of gas and aerosol substances are emitted into the atmosphere. These substances affect significantly the air quality and radiative characteristics of the climatic system (Ito, Ito, & Akimoto, 2007; Lamarque et al., 2010; Rap et al., 2015). Numerous investigations show that the volume of emitted burning products depends significantly on the geographic location of a region, a season, and weather conditions (Ito, Sudo, et al., 2007; Strode & Pawson, 2013; Van Drooge et al., 2016; Vasileva & Moiseenko, 2013; Williamson et al., 2016; Yang et al., 2012). That is why the available estimates of the amount of emitted substances differ widely and are regularly refined (Abbott et al., 2016; Allen, 2016; Schwietzke et al., 2016). It is important to note that year-to-year variations of the area of forest fires and the intensity of atmospheric emissions from them are smaller than the tendency of growing anthropogenic emissions of the analogous compounds (Lamarque et al., 2010).

Highly detailed assessment of chemical composition of the troposphere and yearly burden of emitted atmospheric pollutants can be obtained from ground-based observations. These data allow one to calculate the emission of aerosol and gas compounds and to assess the worsening of air quality. Data from airborne sensing measurements provide spatial and temporal distribution of biomass burning products. However, the method of airborne sensing is characterized by complexity and high costs. That is why such measurements are relatively few (Basso et al., 2016; Bian et al., 2013; Law et al., 2014; O'Shea et al., 2013; Palmer et al., 2013; Virkkula et al., 2014). There is a need to expand the airborne measurements of atmospheric composition study to characterize distribution of biomass burning products emitted by forest fires.

In Russia, seven events of very powerful forest fires were observed in the last decades: in 2002 and 2010 at the European territory, in 2012 in Western and Eastern Siberia including Yakutiya, in 2014–2016 in the Baikal region and Buryatiya. European fires are studied quite thoroughly by now (Chubarova et al., 2011; Elansky et al., 2011; Fokeeva et al., 2011; Kashin et al., 2016; Krol et al., 2013; Vivchar et al., 2010; Zvyagintsev et al., 2011). The analysis of Siberian fires is not complete (Shtabkin et al., 2016; Sklyadneva et al., 2015).

Table 1
Gas Analysis and Aerosol Equipment

Unit	Device/sensor	Parameter	Range	Error	Time constant
Gas analysis system	G2301-m	CO_2, ppm	0, …, 1,000	< 0.2 ppm	1 s
		CH_4, ppm	0, …, 20	< 0.0015 ppm	1 s
		H_2O, ppm	0, …, 7,0000	< 150 ppm	1 s
	TEI Model 49C	O_3, ppm	0, …, 200	0.001 ppm	4 s
	TEI Model 48C	CO, ppm	0, …, 1,000	± 1%	1 s
	LI-6262	CO_2, ppm	0, …, 1,000	< 0.2 ppm[a]	1 s
Aerosol system	GRIMM #1.109	D_p, μm	0.25, …, 32	—	6 s
		(31 hannels) N, cm^{-3}	0, …, 2,000	±3%	
	Diffusional aerosol spectrometer	D_p, nm	3, …, 200	—	80 s
		(20 channels) N, cm^{-3}	0, …, 500,000	±10%	
	FAN nephelometer	σ, km^{-1}	—	—	1 s
	AMA-02 aethalometer	BC, $\mu g/m^3$	0.01, …, 100	0.01 $\mu g/m^3$	20 s
Gammabackground	IRF	γ background, μR/h	1, …, 1,000	30%	1 s

[a]Calibrated every 15 min against reference gas mixtures (NIES scale, National Institute for Environmental Studies).

Therefore, this study focuses on analyses of vertical distribution of aerosol and gas admixtures measured during forest fires in Siberia in 2012.

2. Methods Used and Characterization of the Period Under Study

For the large-scale study of the air composition distribution in the troposphere over Siberia, we used the Optik Tu-134 aircraft laboratory. Its detailed description can be found in Anokhin et al. (2011). The measurements in this experiment were conducted with gas analysis and aerosol measurements described in Table 1.

The airborne study was carried out on 31 July and 1 August 2012 along the route Novosibirsk-Tomsk-Mirnyi-Yakutsk-Bratsk-Novosibirsk and consisted of five parts caused by the need of aircraft refueling in airports of the mentioned cities. During the flights between the cities, the aircraft made several ascents (up to an attitude ≈8.2 km) and descents down to the minimal altitude safe for the horizontal flight. Thus, a total of 20 vertical profiles of the concentrations of greenhouse gases and aerosols were obtained. The flight scheme is shown in Figure 1.

In this period, the large part of the Siberian territory was covered by numerous forest fires, which caused the high level of atmospheric pollution by burning products. The fire situation can be judged from the tabulated and plotted data obtained by the Krasnoyarsk Regional EMERCOM Administration for July 31 and August 1 of 2012 (Table 2 and Figure 2). Figure 2 shows the map of hot spots and smoke plumes from fires (in yellow) recorded for 31 July 2012 (upper panel) and 1 August 2012 (lower panel). Blue line represents the track of the aircraft laboratory flight along the route Novosibirsk-Tomsk-Mirnyi-Yakutsk-Bratsk-Novosibirsk. The

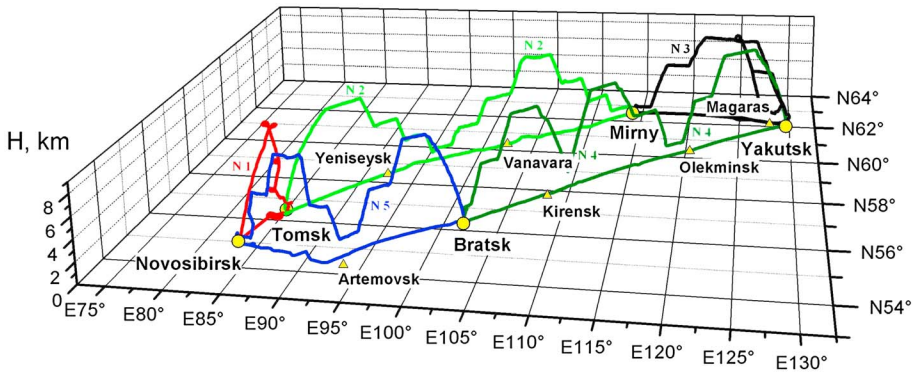

Figure 1. Diagram of atmospheric sensing. Yellow circle dots show the refueling sites.

Table 2
Fire Situation at the Siberian Territory for 31 July and 1 August 2012

Federal District	Subject	Number of thermal points	Number of fires	Area of fire sites, ha	Burning area, ha
		31 July 2012			
Siberian	Novosibirsk Region	2	1	4,258	31,709
Siberian	Tomsk Region	54	21	15,215	145,660
Siberian	Krasnoyarsk Territory	733	220	263,303	2,360,094
Siberian	Irkutsk Region	20	8	14,522	17,722
Far East	Sakha (Yakutiya) Republic	744	164	428,675	1,560,742
		1 August 2012			
Far East	Sakha Republic (Yakutiya)	800	200	531,521	1,847,748
Siberian	Irkutsk Region	29	13	22,659	32,203
Siberian	Krasnoyarsk Territory	274	118	109,395	1,796,093
Siberian	Kakasiya Republic	2	1	309	352
Siberian	Kemerovo Region	1	1	207	558
Siberian	Novosibirsk Region	N/A	N/A	N/A	N/A

largest area covered by wildfires on 31 July (428,675 ha) was reported for Sakha (Yakutia) Republic (bounded by purple lines in the right upper corner of the map, Figure 2a), which was surveyed at the end of the first day of campaign. The most difficult situation with fires had developed over Krasnoyarsk Territory where the maximum number of hot spots was observed. On the next day, burned area over Sakha region extended to the west, while at Krasnoyarsk Territory it shrank. Wildfires in Siberia were observed through the whole summer of 2012 beginning from June. Five days prior to the flight campaign, burning area over Tomsk, Krasnoyarsk, and Yakutia regions was 749925 ha, 2,181,559 ha, and 707,723 ha, respectively.

The synoptic situation during the experiment can be judged from maps shown in Figure 3.

The analysis of the synoptic situation (Figure 3) shows that the experiment was mostly conducted under conditions of the low-gradient field (calm weather) that led to the accumulation of pollutants in the region, and dry lightning aggravated the situation. We have found no data about the influence of fires in 2012 on other regions of Russia, whereas the traces of fires of 2014 and 2015 were found at large distances, in other countries (Jung et al., 2016; Laing et al., 2016).

This can be clearly seen from the satellite data (http://giovanni.sci.gsfc.nasa.gov) of the total content of carbon monoxide and aerosols (aerosol optical depth) shown in Figure 4.

3. Vertical Distribution of Carbonaceous Gases

Figure 5 depicts the vertical profiles of the CO_2, CH_4, and CO concentrations obtained during the airborne experiment over the Siberian territory from Novosibirsk to Yakutsk in summer 2012. The vertical distribution of the concentrations of these gases during the 2012 summer campaign differs widely from the earlier campaigns. The fires have led to the situation that many profiles are characterized by the presence of layers with concentrations far exceeding the background levels as a result of intersection of smoke plumes by the aircraft.

The vertical profiles shown in Figure 5 differ markedly from both the average (Miyamoto et al., 2013) and model profiles (Saito et al., 2013; Shirai et al., 2012; Xueref-Remy et al., 2011), as well as from profiles measured in the absence of forest fires (Andreae et al., 2012; Beck et al., 2012; Biraud et al., 2013; Gatti et al., 2010; Girach & Nair, 2014; Paris et al., 2009, 2010; Xiong et al., 2010). Next, we discuss details of observed variability in CO, CO_2, and CH_4 profiles with regard to the age of the smoke transported from the fire sources.

The situation with forest fires in the Tomsk region in summer 2012 was severe, and this has led to heavy smoke presence over Tomsk in the period of 24–28 July. Before the start of airborne sensing, in connection with rains and alternation of the wind direction on 28 July, the atmosphere over Tomsk became more clean if we look at CO profile. That is why the concentrations of all the three components were close to the background values. The sole exception was a thin smoke layer observed during the landing in the Bogashevo Airport. This plume can be considered as residual (aged), because it was clearly pronounced only in increase of the CO concentration up to 450 ppb. "Younger" smokes are characterized by high concentrations of not only CO but also CO_2 and CH_4 simultaneously. In turn, the correlation between these concentrations

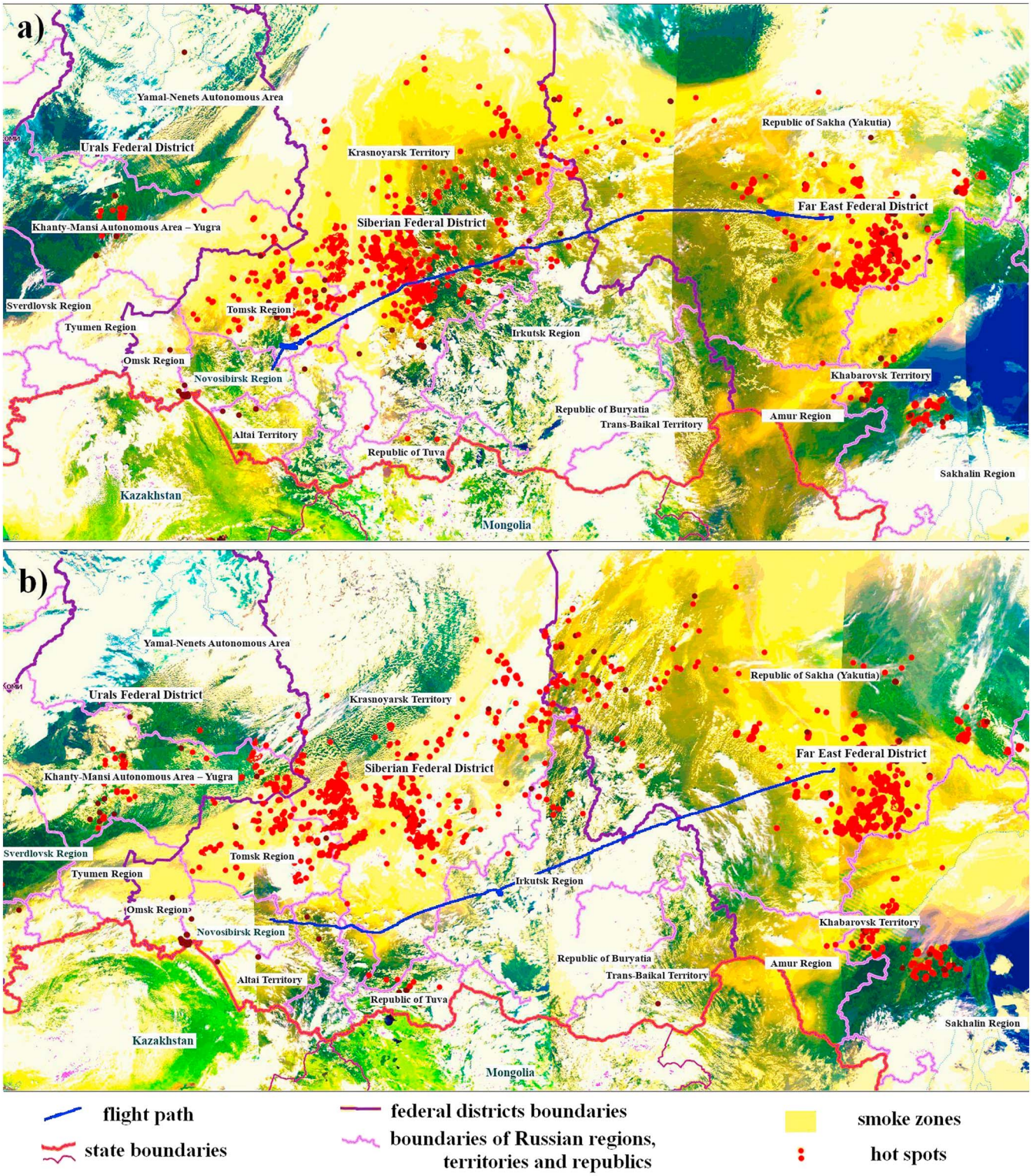

Figure 2. Map of hot spots and smokes from fires: (a) 31 July 2012; (b) 1 August 2012. Blue line shows the flight of the aircraft laboratory along the route Novosibirsk-Tomsk-Mirnyi-Yakutsk-Bratsk-Novosibirsk.

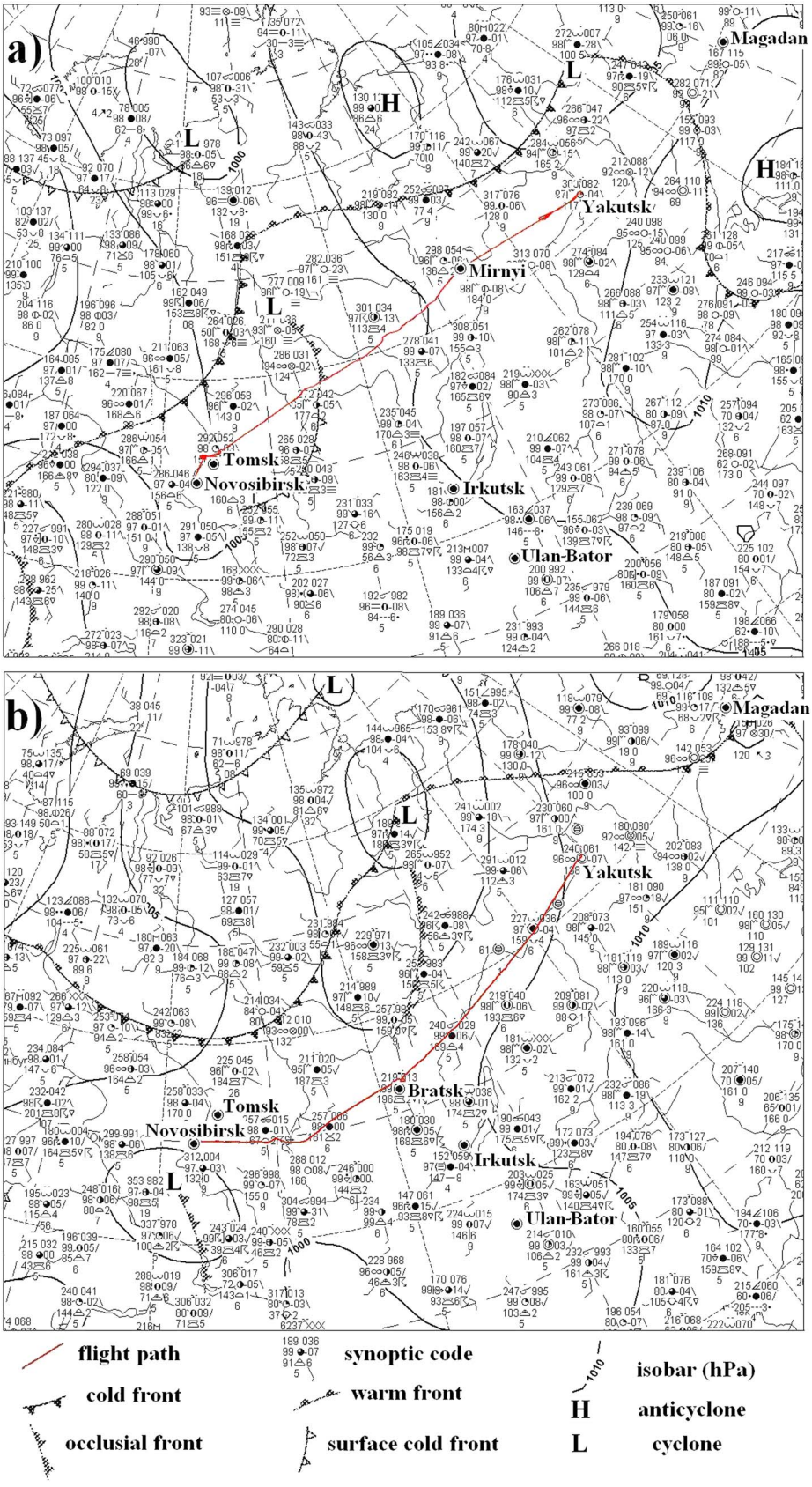

Figure 3. Near-ground weather maps: (a) 31 July 2012; (b) 1 August 2012.

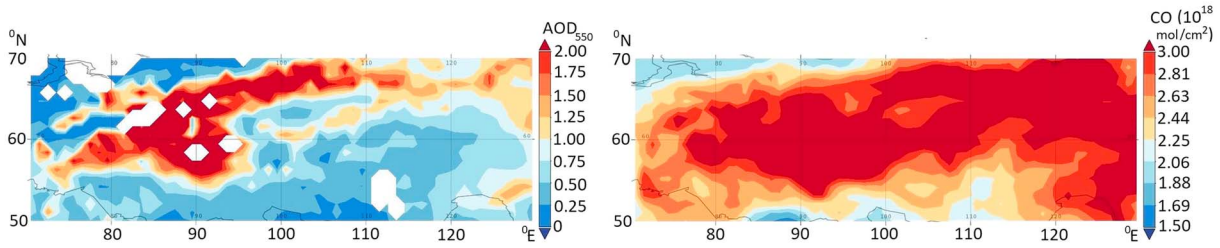

Figure 4. Spatial distribution of aerosol optical depth (AOD) and carbon monoxide (CO) in 29 July to 3 August 2012.

allows us to identify whether an increase of the methane concentration is a consequence of emissions from biomass burning or long-range transport, for example, from wetlands.

The highest levels of all the three studied gases during this flight campaign were observed not only in the lower troposphere (<1.5 km) but also in the free atmosphere at heights of 4–5 km. This fact indicates that the gases penetrated above the boundary layer.

The maximal one-time concentrations of CO_2, CH_4, and CO over fires achieved 432 ppm, 2367 ppb, and 4036 ppb, respectively. In addition to this layer, several layers with the increased concentrations of CO_2, CH_4, and CO were observed in this profile at heights of 2.5, 3.5, and 5.2 km in elevated smoke plumes.

At the third part of the route (N 3 in Figure 1), when approaching Yakutsk, one more layer with the very high one-time concentrations of CO_2, CH_4, and CO (up to 4,12 ppm, 2,139 ppb, and 2,606 ppb, respectively) with

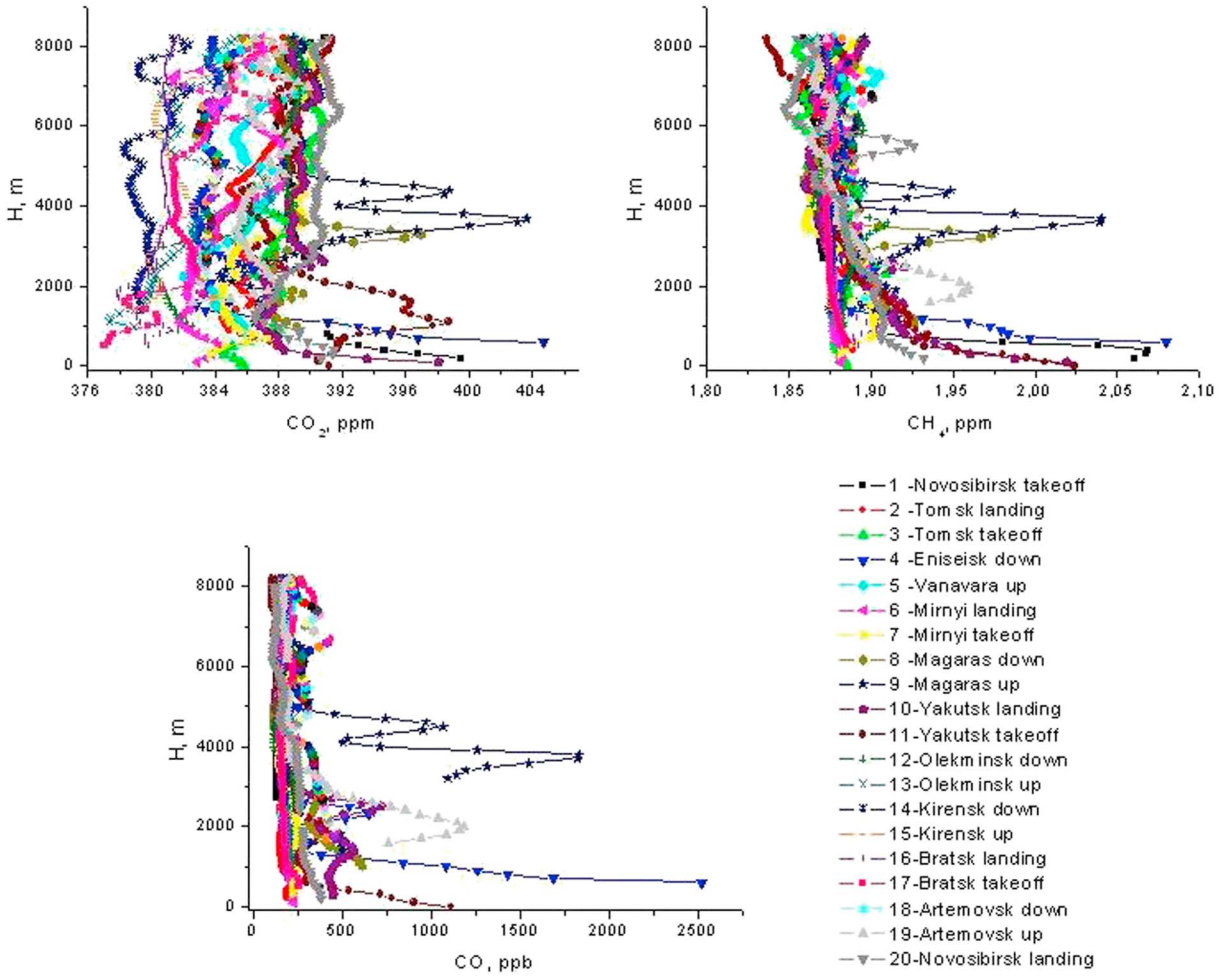

Figure 5. Vertical profiles of the CO_2, CH_4, and CO concentrations obtained in the flights of the 2012 campaign (averaged by smoothing in steps of 100 m on height).

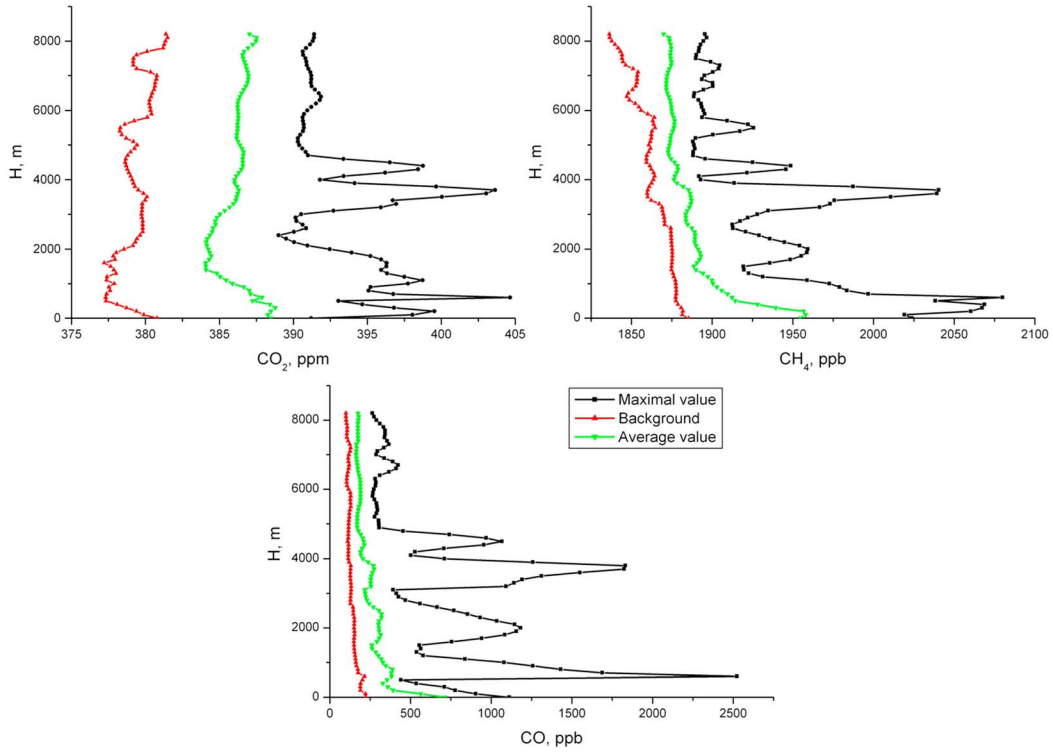

Figure 6. Vertical profiles of the background, average, and maximal concentrations of CO_2, CH_4, and CO at the route Novosibirsk-Yakutsk-Novosibirsk.

the vertical thickness from 2 to 5 km was observed in the free troposphere. At the landing in Yakutsk, the concentrations were also high, since fires were located in close vicinity to Yakutsk.

For the night, the situation in Yakutsk did not improve, and the concentrations in the lower part of the boundary layer remained at the level of the previous day, except for CO_2, a part of which was likely already (at 13:28 local time) assimilated by plants untouched by fires. After takeoff, during the climb, the aircraft intersected one fresher smoke plume in the layer of 1.2–2.2 km. Then, the flight to Bratsk was mostly carried out under the background conditions except for several cases of passage of old diluted plumes at different heights in the free troposphere, because the most significant increases were seen only in the concentrations of CH_4 and CO.

If we consider the CO_2 profiles, then, if smoke plumes are excluded from the consideration, we can notice the difference in the background values observed in the troposphere over the Irkutsk Region and regions covered by fires (profiles 1–12 and 19–20), which amounted to ≈5 ppm. Most probably, this difference is connected with accumulation of carbon dioxide emitted by forest fires in the troposphere over fire regions under conditions of abnormally high air temperatures observed in Siberia in summer 2012 for a long period along with the absence of considerable precipitations.

The measured concentrations of carbonaceous gases over the territory of Western and Eastern Siberia are quite comparable with or higher than the analogous data for the vertical distributions in other regions (Basso et al., 2016; Bian et al., 2013; Law et al., 2014; O'Shea et al., 2013; Palmer et al., 2013; Virkkula et al., 2014). At the same time, the measured maximal concentrations of CO in the troposphere are smaller than those measured in the surface layer (Elansky et al., 2011; Fokeeva et al., 2011; Kashin et al., 2016; Krol et al., 2013; Zvyagintsev et al., 2011), except for CO_2. Probably, in our case this is caused by the fact that the aircraft flew over the territory occupied by boreal forests.

To estimate the emission of carbonaceous gases, some authors calculate the enrichment coefficients or emission factors (Cordell et al., 2016; Garcia-Hurtado et al., 2013; Huijnen et al., 2016; Liu et al., 2016; Meyer et al., 2012; Surawski et al., 2016). Due to synoptic conditions in the period of our experiment, the vertical profiles

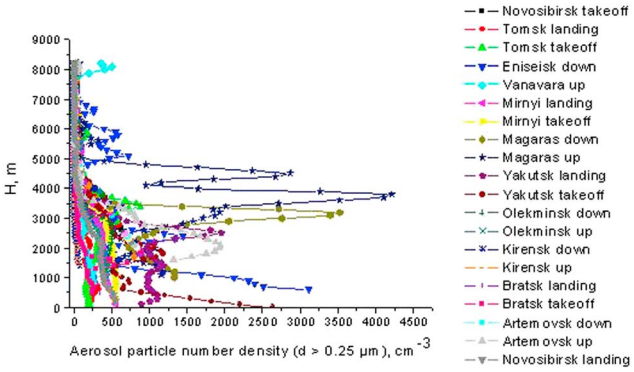

Figure 7. Aerosol particle number density ($d > 0.25$ μm) along the route Novosibirsk-Yakutsk-Novosibirsk.

measured over the region are similar due to mixing of the pollution from the smoke under low wind conditions (Figure 5). Thus, we have constructed average profiles and also determined the maximum values of the concentrations of carbon gases for flight. If we take the similar profiles from the previous summer campaign along the same route, then we can find the maximal and average increment of the concentrations of carbonaceous gases. These data are shown in Figure 6.

It can be seen from Figure 6 that for CO_2 the increment of the concentration in the zone of action of forest fires varies with height. On average, it is 10.5 ppm in the boundary layer and 5.6 ppm in the free atmosphere. The maximal increment can be 27 ppm in the boundary layer and 24 ppm in the free atmosphere.

For methane, the average increment varies from 75 ppb in the boundary layer to 30 ppb in the upper troposphere. The increment in the midtroposphere (3–6 km) appeared to be smaller (30 ppb) than in the upper troposphere. The maximal concentrations of CH_4 can increase from 202 ppb in the boundary layer to 180 ppb in the midtroposphere and 50–60 ppb in the upper troposphere.

For carbon monoxide, the average and maximal increments differ by an order of magnitude. Thus, in the boundary layer, the maximal difference can achieve 2,300 ppb at the average concentration of 170 ppb. In the layers located in the midtroposphere, the maximal increment can be 1,000 and 1,700 ppb, whereas its average values range within 150–120 ppb. For CO_2 and CH_4, the obtained estimates are comparable with the data of Basso et al. (2016), Bian et al. (2013), Law et al. (2014), O'Shea et al. (2013), Palmer et al. (2013), and Virkkula et al. (2014), while for CO they exceed the published data.

4. Vertical Distribution of Aerosol

Biomass burning emits not only carbonaceous gases but also a large amount of aerosol particles, first of all, of the fine and submicron fractions (Popovicheva et al., 2016; Rakhimov et al., 2014). The vertical distribution of the aerosol particle number density ($d > 0.25$ μm) over different Siberian regions is shown in Figure 7.

It can be seen from Figure 7 that the aerosol particle number density in plumes achieved 4,400 cm^{-3}. Outside the plumes, the number density varied within 400–1,000 cm^{-3} depending on a region. To be noted is the presence of plumes in the middle troposphere, that is, above the boundary layer.

The shapes of the depicted profiles differ significantly from those obtained under the background conditions in the absence of emission plumes (Hamburger et al., 2012; Matsui et al., 2011). It is difficult to compare the absolute values of the number densities, because different investigators use different size ranges of particles measured in experiments.

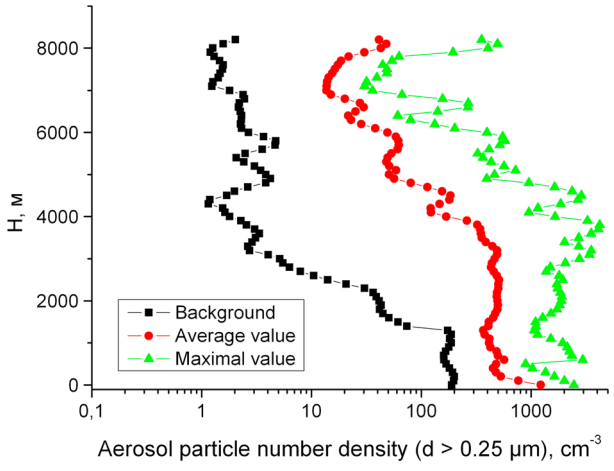

Figure 8. Background, average, and maximal number densities at the route (log scale).

Analogously to the carbonaceous gases, the average and the maximal profiles were calculated from the data of all ascents and descends of the entire route. The data obtained are shown in Figure 8.

The background profile is close to those obtained in the absence of forest fire products by other authors (Hamburger et al., 2012; Matsui et al., 2011). In comparison with the background conditions, the average aerosol particle number density increases in the atmosphere subject to forest fire emissions 2.5 times in the boundary layer and 500 times in the middle troposphere and 20–30 times in the upper troposphere. The maximal excess can achieve 15 times in the boundary layer, 4,000 times in the midtroposphere, and 300 times in the upper troposphere.

In addition to the particle number density, the chemical composition of aerosol was determined during the flights. According to Calvo et al. (2013), the chemical composition allows one to assess, in the first approximation, the source of aerosol particles. Particulate matter was collected

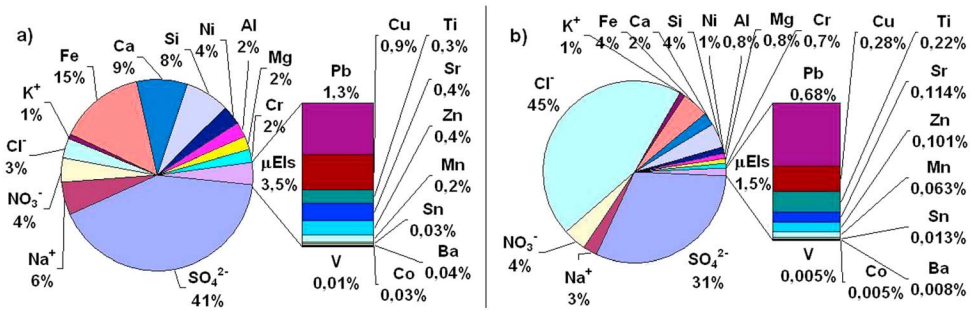

Figure 9. Chemical composition of aerosol: (a) background; (b) plume of forest fire.

onto filters of AFA type (Petryanov's analytical aerosol filters). The chemical analysis of the samples was carried out in the analytical laboratory of the Tomsk State University.

Figure 9 demonstrates the quite significant differences in the chemical composition of aerosol in the background air and in the plume. Both quantitative and qualitative changes can be noticed. The quantitative changes are clearly seen from Table 3.

It follows from Table 3 that the identified mass of aerosol increased almost 7 times (6.9) in the plume. The concentrations of some elements and ions in the plume with respect to the background were enriched from 1.3 to 9.1 times.

Similar results were also obtained by other authors both for the concentrations and for the set of elements and ions (Zvyagintsev et al., 2010; Snider et al., 2016).

The sole exception is Cl^- ion, whose content in the plume is high and the contribution to the total concentration is high too. In other cases, investigators, to the contrary, more often noticed the deficit of chlorine (Lanz et al., 2010; Sarin et al., 2010).

This result is not random. Similar results were obtained in the Norilsk region, as well as in the plume of a forest fire (Arshinov et al., 2006). Now this fact can hardly be explained. At least, so high concentrations were not observed in emissions of industrial enterprises (Belan et al., 1996). Possibly, it is caused by chlorine accumulation in plants as a result of activity of some industries.

Table 3
Values of Elemental and Ionic Composition of Atmospheric Aerosol ($\mu g/m^3$; With Standard Errors Due To Methods of Analysis) and Their Ratios in the Forest Fire Plume and in the Background Region

Component (element or ion)	Plume value ± std. error	Background ± std. error	Plume/ background ratio
Al	0.331 ± 0.066	0.143 ± 0.029	2.3
Ba	0.0033 ± 0.0007	0.0026 ± 0.0005	1.3
Ca	0.935 ± 0.187	0.499 ± 0.100	1.9
Cu	0.111 ± 0.022	0.050 ± 0.010	2.2
Fe	1.786 ± 0.357	0.841 ± 0.168	2.1
Mg	0.306 ± 0.061	0.118 ± 0.024	2.6
Mn	0.025 ± 0.005	0.010 ± 0.002	2.5
Ni	0.475 ± 0.095	0.203 ± 0.041	2.3
Pb	0.270 ± 0.054	0.074 ± 0.015	3.6
Sn	0.0053 ± 0.0011	0.0018 ± 0.0004	2.9
Ti	0.086 ± 0.017	0.019 ± 0.004	4.4
V	0.0020 ± 0.0004	0.0006 ± 0.00012	3.1
Si	1.793 ± 0.359	0.435 ± 0.087	4.1
Co	0.0020 ± 0.0004	0.0016 ± 0.0003	1.3
Cr	0.283 ± 0.057	0.139 ± 0.028	2.0
Sr	0.045 ± 0.009	0.024 ± 0.005	1.9
Zn	0.040 ± 0.008	0.021 ± 0.004	1.9
Na^+	1.055 ± 0.106	0.331 ± 0.033	3.2
K^+	0.533 ± 0.053	0.059 ± 0.006	9.1
Cl^-	17.873 ± 2.144	0.184 ± 0.022	97.3
NO_3^-	1.670 ± 0.167	0.239 ± 0.024	7.0
SO_4^{2-}	12.306 ± 0.984	2.388 ± 0.191	5.2
Total	39.936 ± 4.752	5.783 ± 0.792	6.9

5. Vertical Distribution of Ozone

The vertical ozone distribution under conditions of forest fires differs drastically from profiles of carbonaceous gases and aerosol (Figure 10). First, the common feature for almost all profiles is the increase of concentration with height. Second, they are characterized by the wide spread in the content values depending on the part of the route. A close result was also obtained in Parrington et al. (2013).

This distribution is explained by the fact that under conditions of a high aerosol load a strong deposition of ozone on aerosol particle occurs (Belan et al., 1992; Belan, 2010). Due to reducing UV-B radiation intensity in the layer of emission products, the ozone generation in the troposphere decreases.

Despite the presence of sink, it can be seen from Figure 10 that the ozone concentration is increased in some layers. This fact indicates that the photochemical generation of ozone from products of forest fires proceeds even under decreased power of UV-B radiation in the troposphere. The fact of ozone generation in the surface air layer was already noted in the literature (Jaffe & Wigder, 2012; Jena et al., 2015; Wigder et al., 2013).

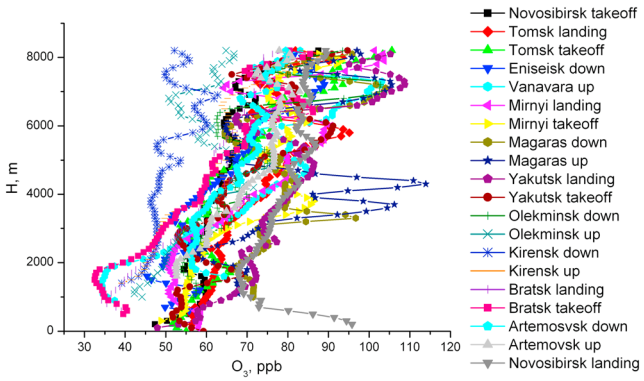

Figure 10. Vertical ozone distribution along the route Novosibirsk-Yakutsk-Novosibirsk.

The data of our measurements have shown that the ozone is generated from biomass burning products at the boundary of an emission plume. Two versions of this process are possible: generation above the plume and generation both above and under the plume. The type of the process is independent of the relative position of the layer. As an illustration, Figure 11 shows fragments of profiles near Yakutsk and Magaras, when two plumes with different types of ozone generation were observed in each flight.

It can be seen from Figure 11a, which also shows the data on the number density of carbon monoxide, that two plumes were observed over Yakutsk: one with the maximum near 800 m and another with the maximum near 1,900 m. Two maxima of the ozone concentration were pronounced above and under the first plume. In the second case, the ozone maximum was observed above the upper plume. Over Magaras, the situation was contrary. One ozone maximum was observed under the lower plume and two maxima were associated with the upper plume. It is likely that two plumes have been transported with air masses of different age, because it is known that aged plumes have enhanced levels of ozone. Also, it is might be that enhanced ozone levels was a result of the strong convection over intense fires that tend to create pyrocumulous clouds and associated rapid cooling of the lifted air masses that lead to the strong downdraft and stratosphere/troposphere exchange (Fromm et al., 2010; Trentmann, Andreae, & Graf, 2003; Trentmann, Fruh, et al., 2003). There is also a slight likelihood that some fire plumes has been mixed with air masses affected by anthropogenic pollution, for example, the presence of NO_x in the urban plums may influence the ozone chemistry and therefore its production (Singh et al., 2012).

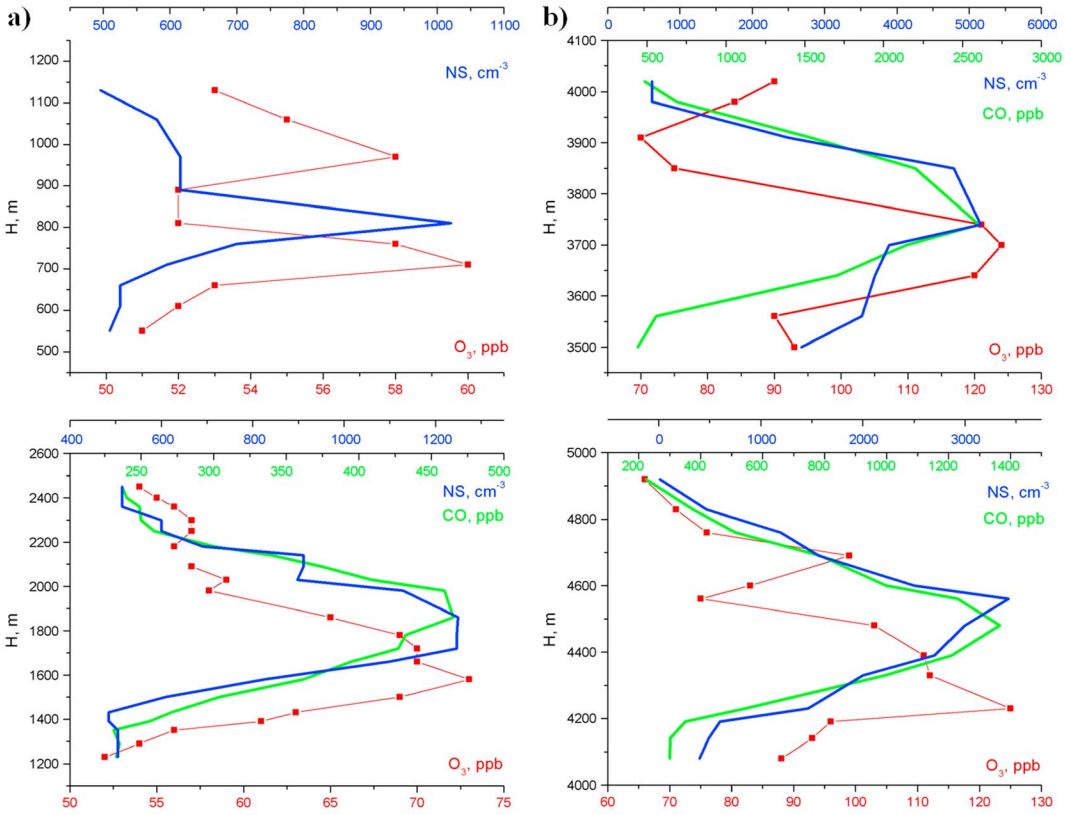

Figure 11. Vertical distribution of ozone (O_3), carbon monoxide (CO), and aerosol number density (NS) over Yakutsk (a) and Magaras (b).

6. Conclusions

Thus, this study has shown that emissions of biomass burning products from forest fires under conditions of the low-gradient field (calm weather) at the low diffusion in the atmosphere lead to accumulation of pollutants in the lower troposphere and their partial inflow to the upper troposphere. The highest levels of all the three studied gases during this flight campaign were observed not only in the lower troposphere (<1.5 km) but also in the free atmosphere at heights of 4–5 km. This fact indicates that the gases penetrated above the boundary layer.

The accumulation of pollutants causes a significant increase in the concentration of carbonaceous gases in the lower and middle troposphere and a multiple increase in the number density and mass concentration of aerosol. The maximal concentrations of CO_2, CH_4, and CO over fires achieved 432 ppm, 2,367 ppb, and 4,036 ppb, respectively. In addition to this layer, several layers with the increased concentrations of CO_2, CH_4, and CO were observed in this profile at heights of 2.5, 3.5, and 5.2 km in elevated smoke plumes. The aerosol particle number density in plumes achieved 4,400 cm^{-3}. Outside the plumes, the number density varied within 400–1,000 cm^{-3} depending on a region. To be noted is the presence of plumes in the middle troposphere, that is, above the boundary layer. The average aerosol particle number density increases in the atmosphere subject to forest fire emissions 2.5 times in the boundary layer and 500 times in the middle troposphere and 20–30 times in the upper troposphere. The maximal excess can achieve 15 times in the boundary layer, 4,000 times in the midtroposphere, and 300 times in the upper troposphere. As this takes place, the amounts of elements and ions in the aerosol composition increase selectively. The identified mass of aerosol increased almost 7 times (6.9) in the plume. The concentrations of some elements and ions in the plume with respect to the background were enriched from 1.3 (Ba, Co) to 9.1 (K^+) times.

The vertical distribution of ozone has some peculiarities. The ozone concentration decreases in the layers, where aerosol is accumulated, and increases in the absence of aerosol particles. At the same time, photochemical generation of ozone is observed at the boundaries of fire plumes in the zone of entrainment of fresh air.

Acknowledgments
This study was supported by Russian Science Foundation, 17-17-01095. Access to the data provided in article is organized on the website of the Russian-French YAK-AEROSIB project to the address: https://yak-aerosib.lsce.ipsl.fr/doku.php?

References

Abbott, B. W., Jones, J. B., Schuur, G., Chapin, E. A. III, Bowden, F. S., Bret-Harte, W. B., et al. (2016). Biomass offsets little or none of permafrost carbon release from soils, streams, and wildfire: An expert assessment. *Environmental Research Letters*, *11*(3), 034014. https://doi.org/10.1088/1748-9326/11/3/034014

Allen, G. (2016). Rebalancing the global methane budget. *Nature*, *538*(7623), 46–48. https://doi.org/10.1038/538046a

Andreae, M. O. (1991). Biomass burning: Its history, use and distribution and its impact on environmental quality and global climate. In J. S. Levine (Ed.), *Global biomass burning: Atmospheric, climatic and biospheric implications* (pp. 3–21). Cambridge, MA: MIT Press.

Andreae, M. O., Artaxo, P., Beck, V., Bela, M., Freitas, S., Gerbig, C., et al. (2012). Carbon monoxide and related trace gases and aerosols over the Amazon Basin during the wet and dry seasons. *Atmospheric Chemistry and Physics*, *12*(13), 6041–6065. https://doi.org/10.5194/acp-12-6041-2012

Anokhin, G. G., Antokhin, P. N., Arshinov, M. Y., Barsuk, V. E., Belan, B. D., Belan, S. B., et al. (2011). OPTIK Tu-134 aircraft laboratory. *Atmospheric and Oceanic Optics*, *24*(9), 805–816.

Arshinov, M. Y., Belan, B. D., Ivlev, G. A., Krasnov, O. A., Rasskazchikova, T. M., Simonenkov, D. V., et al. (2006). Complex assessment of the conditions of the air basin over Norilsk industrial region. Part 3. Dispersal of admixtures. *Atmospheric and Oceanic Optics*, *19*(9), 717–723.

Basso, L. S., Gatti, L. V., Gloor, M., Miller, J. B., Domingues, L. G., Correia, C. S. C., & Borges, V. F. (2016). Seasonality and interannual variability of CH_4 fluxes from the eastern Amazon Basin inferred from atmospheric mole fraction profiles. *Journal of Geophysical Research: Atmospheres*, *121*, 168–184. https://doi.org/10.1002/2015JD023874

Beck, V., Chen, H., Gerbig, C., Bergamaschi, P., Bruhwiler, L., Houweling, S., et al. (2012). Methane airborne measurements and comparison to global models during BARCA. *Journal of Geophysical Research*, *117*, D15310. https://doi.org/10.1029/2011JD017345

Belan, B. D. (2010). Tropospheric ozone. 7. Sinks of ozone in troposphere. *Optika Atmosfery i Okeana*, *23*(2), 108–126.

Belan, B. D., Panchenko, M. V., Terpugova, S. A., & Tolmachev, G. N. (1992). Estimation of the ozone sink on aerosol particles. *Atmospheric and Oceanic Optics*, *5*(6), 409–411.

Belan, B. D., Simonenkov, D. V., & Tolmachev, G. N. (1996). Chemical composition of industrial aerosol in some regions. *Atmospheric and Oceanic Optics*, *9*(4), 287–292.

Bian, H., Colarco, P. R., Chin, M., Chen, G., Rodriguez, J. M., Liang, Q., et al. (2013). Source attributions of pollution to the Western Arctic during the NASA ARCTAS field campaign. *Atmospheric Chemistry and Physics*, *13*(9), 4707–4721. https://doi.org/10.5194/acp-13-4707-2013

Biraud, S. C., Torn, M. S., Smith, J. R., Sweeney, C., Riley, W. J., & Tans, P. P. (2013). A multi-year record of airborne CO_2 observations in the US Southern Great Plain. *Atmospheric Measurement Techniques*, *6*(3), 751–763. https://doi.org/10.5194/amt-6-751-2013

Calvo, A. I., Alves, C., Castro, A., Pont, V., Vicente, A. M., & Fraile, R. (2013). Research on aerosol sources and chemical composition: Past, current and emerging issues. *Atmospheric Research*, *120-121*, 1–28. https://doi.org/10.1016/j.atmosres.2012.09.021

Chubarova, N. E., Gorbarenko, E. V., Nezval', E. I., & Shilovtseva, O. A. (2011). Aerosol and radiation characteristics of the atmosphere during forest and peat fires in 1972, 2002, and 2010 in the region of Moscow, *Izvestiya*. *Atmospheric and Oceanic Physics*, *47*(6), 729–738. https://doi.org/10.1134/S0001433811060028

Cordell, R. L., Mazet, M., Dechoux, C., Hama, S. M. L., Staelens, J., Hofman, J., et al. (2016). Evaluation of biomass burning across North West Europe and its impact on air quality. *Atmospheric Environment*, *141*, 276–286. https://doi.org/10.1016/j.atmosenv.2016.06.065

Elansky, N. F., Mokhov, I. I., Belikov, I. B., Berezina, E. V., Elokhov, A. S., Ivanov, V. A., et al. (2011). Gaseous admixtures in the atmosphere over Moscow during the 2010 summer, *Izvestiya*. *Atmospheric and Oceanic Physics*, *47*(6), 672–681. https://doi.org/10.1134/S000143381106003X

Fokeeva, E. V., Safronov, A. N., Rakitin, V. S., Yurganov, L. N., Grechko, E. I., & Shumskii, R. A. (2011). Investigation of the 2010 July–August fires impact on carbon monoxide atmospheric pollution in Moscow and its outskirts, estimating of emissions, *Izvestiya*. *Atmospheric and Oceanic Physics*, *47*(6), 682–698. https://doi.org/10.1134/S0001433811060041

Fromm, M., Lindsey, D. T., Servranckx, R., Yue, G., Trickl, T., Sica, R., et al. (2010). The untold story of pyrocumulonimbus. *Bulletin of the American Meteorological Society*, *91*(9), 1193–1210. https://doi.org/10.1175/2010BAMS3004.1

Garcia-Hurtado, E., Pey, J., Baeza, M. J., Carrara, A., Llovet, J., Querol, X., et al. (2013). Vallejo carbon emissions in Mediterranean shrubland wild fires: An experimental approach. *Atmospheric Environment*, *69*, 86–93. https://doi.org/10.1016/j.atmosenv.2012.11.063

Gatti, L. V., Miller, J. B., D'Amelio, M. T. S., Martinewski, A., Basso, L. S., Gloor, M. E., et al. (2010). Vertical profiles of CO_2 above eastern Amazonia suggest a net carbon flux to the atmosphere and balanced biosphere between 2000 and 2009. *Tellus*, *62*(5), 581–594. https://doi.org/10.1111/j.1600-0889.2010.00484.x

Girach, I. A., & Nair, P. R. (2014). On the vertical distribution of carbon monoxide over Bay of Bengal during winter: Role of water vapour and vertical updrafts. *Journal of Atmospheric and Solar-Terrestrial Physics*, *117*, 31–47. https://doi.org/10.1016/j.jastp.2014.05.003

Hamburger, T., Meeking, G. M., Minikin, A., Petzold, A., Coe, H., & Krejci, R. (2012). Airborne observations of aerosol microphysical properties and particle ageing processes in the troposphere above Europe. *Atmospheric Chemistry and Physics*, *12*(23), 11,533–11,554. https://doi.org/10.5194/acp-12-11533-2012

Huijnen, V., Wooster, M. J., Kaiser, J. W., Gaveau, D. L. A., Flemming, J., Parrington, M., et al. (2016). Fire carbon emissions over maritime southeast Asia in 2015 largest since 1997. *Scientific Reports*, *6*(1), 8. https://doi.org/10.1038/srep26886

Ito, A., Ito, A., & Akimoto, H. (2007). Seasonal and interannual variations in CO and BC emissions from open biomass burning in Southern Africa during 1998–2005. *Global Biogeochemical Cycles*, *21*, GB2011. https://doi.org/10.1029/2006GB002848

Ito, A., Sudo, K., Akimoto, H., Sillman, S., & Penner, J. E. (2007). Global modeling analysis of tropospheric ozone and its radiative forcing from biomass burning emissions in the twentieth century. *Journal of Geophysical Research*, *112*, D24307. https://doi.org/10.1029/2007JD008745

Jaffe, D. A., & Wigder, N. L. (2012). Ozone production from wild fires: A critical review. *Atmospheric Environment*, *51*(1), 1–10. https://doi.org/10.1016/j.atmosenv.2011.11.063

Jena, C., Ghude, S. D., Pfister, G. G., Chate, D. M., Kumar, R., Beig, G., et al. (2015). Influence of springtime biomass burning in South Asia on regional ozone (O3): A model based case study. *Atmospheric Environment*, *100*, 37–47. https://doi.org/10.1016/j.atmosenv.2014.10.027

Jung, J., Lyu, H., Lee, M., Hwang, T., Lee, S., & Oh, S. (2016). Impact of Siberian forest fires on the atmosphere over the Korean Peninsula during summer 2014. *Atmospheric Chemistry and Physics*, *16*(11), 6757–6770. https://doi.org/10.5194/acp-16-6757-2016

Kashin, F. V., Aref'ev, V. N., Sizov, N. I., Akimenko, R. M., & Upenek, L. B. (2016). Background component of carbon oxide concentrations in the surface air (Obninsk monitoring station), *Izvestiya*. *Atmospheric and Oceanic Physics*, *52*(3), 247–252. https://doi.org/10.1134/S0001433816030051

Krol, M., Peters, W., Hooghiemstra, P., George, M., Clerbaux, C., Hurtmans, D., et al. (2013). How much CO was emitted by the 2010 fires around Moscow? *Atmospheric Chemistry and Physics*, *13*(9), 4737–4747. https://doi.org/10.5194/acp-13-4737-2013

Laing, J. R., Jaffe, D. A., & Hee, J. R. (2016). Physical and optical properties of aged biomass burning aerosol from wildfires in Siberia and the Western USA at the Mt. Bachelor Observatory. *Atmospheric Chemistry and Physics*, *16*(23), 15,185–15,197. https://doi.org/10.5194/acp-16-15185-2016

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., et al. (2010). Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application. *Atmospheric Chemistry and Physics*, *10*(15), 7017–7039. https://doi.org/10.5194/acp-10-7017-2010

Lanz, V. A., Prevot, A. S. H., Alfarra, M. R., Weimer, S., Mohr, C., DeCarlo, P. F., et al. (2010). Characterization of aerosol chemical composition with aerosol mass spectrometry in Central Europe: An overview. *Atmospheric Chemistry and Physics*, *10*(21), 10,453–10,471. https://doi.org/10.5194/acp-10-10453-2010

Law, K. S., Stohl, A., Quinn, P. K., Brock, C. A., Burkhart, J. F., Paris, J.-D., et al. (2014). Arctic air pollution. New Insights from POLARCAT-IPY. *BAMS*, *95*(12), 1873–1895. https://doi.org/10.1175/BAMS-D-13-00017.1

Liu, X., Huey, L. G., Yokelson, R. J., Wang, Y., Jimenez, J. L., Campuzano-Jost, P., et al. (2016). Agricultural fires in the southeastern U.S. during SEAC^{4}RS: Emissions of trace gases and particles and evolution of ozone, reactive nitrogen, and organic aerosol. *Journal of Geophysical Research: Atmospheres*, *121*, 7383–7414. https://doi.org/10.1002/2016JD025040

Matsui, H., Kondo, Y., Moteki, N., Takegawa, N., Sahu, L. K., Koike, M., et al. (2011). Accumulation-mode aerosol number concentrations in the Arctic during the ARCTAS aircraft campaign: Long-range transport of polluted and clean air from the Asian continent. *Journal of Geophysical Research*, *116*, D20217. https://doi.org/10.1029/2011JD016189

Meyer, C. P., Cook, G. D., Reisen, F., Smith, T. E. L., Tattaris, M., Russell-Smith, J., et al. (2012). Direct measurements of the seasonality of emission factors from savanna fires in northern Australia. *Journal of Geophysical Research*, *117*, D20305. https://doi.org/10.1029/2012JD017671

Miyamoto, Y., Inoue, M., Morino, I., Uchino, O., Yokota, T., Machida, T., et al. (2013). Atmospheric column-averaged mole fractions of carbon dioxide at 53 aircraft measurement sites. *Atmospheric Chemistry and Physics*, *13*(10), 5265–5275. https://doi.org/10.5194/acp-13-5265-2013

O'Shea, S. J., Allen, G., Gallagher, M. W., Bauguitte, S. J.-B., Illingworth, S. M., Le Breton, M., et al. (2013). Airborne observations of trace gases over boreal Canada during BORTAS: Campaign climatology, air mass analysis and enhancement ratios. *Atmospheric Chemistry and Physics*, *13*(24), 12,451–12,467. https://doi.org/10.5194/acp-13-12451-2013

Palmer, P. I., Parrington, M., Lee, J. D., Lewis, A. C., Rickard, A. R., Bernath, P. F., et al. (2013). Quantifying the impact of BOReal forest fires on Tropospheric oxidants over the Atlantic using Aircraft and Satellites (BORTAS) experiment: Design, execution and science overview. *Atmospheric Chemistry and Physics*, *13*(13), 6239–6261. https://doi.org/10.5194/acp-13-6239-2013

Paris, J.-D., Stohl, A., Nédélec, P., Arshinov, M. Y., Panchenko, M. V., Shmargunov, V. P., et al. (2009). Wildfire smoke in the Siberian Arctic in summer: Source characterization and plume evolution from airborne measurements. *Atmospheric Chemical Physics*, *9*(23), 9315–9327. https://doi.org/10.5194/acp-9-9315-2009

Paris, J.-D., Ciais, P., Nedelec, P., Stohl, A., Belan, B. D., Arshinov, M. Y., et al. (2010). New insights on the chemical composition of the Siberian air shed from the YAK AEROSIB aircraft campaigns. *Bulletin of the American Meteorological Society*, *91*(5), 625–642. https://doi.org/10.1175/2009BAMS2663.1

Parrington, M., Palmer, P. I., Lewis, A. C., Lee, J. D., Rickard, A. R., Di Carlo, P., et al. (2013). Ozone photochemistry in boreal biomass burning plumes. *Atmospheric Chemistry and Physics*, *13*(15), 7321–7341. https://doi.org/10.5194/acp-13-7321-2013

Popovicheva, O. B., Kozlov, V. S., Rakhimov, R. F., Shmargunov, V. P., Kireeva, E. D., Persiantseva, N. M., et al. (2016). Optical-microphysical and physical-chemical characteristics of Siberian biomass burning: Small-scale fires in an aerosol chamber. *Atmospheric and Oceanic Optics*, *29*(6), 492–500. https://doi.org/10.1134/S1024856016060129

Rakhimov, R. F., Kozlov, V. S., Panchenko, M. V., Tumakov, A. G., & Shmargunov, V. P. (2014). Properties of atmospheric aerosol in the plumes of forest fires according to the spectronephelometric measurements. *Atmospheric and Oceanic Optics*, *27*(3), 275–282. https://doi.org/10.1134/S1024856014030075

Rap, A., Spracklen, D. V., Mercado, L., Reddington, C. L., Haywood, J. M., Ellis, R. J., et al. (2015). Fires increase Amazon forest productivity through increases in diffuse radiation. *Geophysical Research Letters*, *42*, 4654–4662. https://doi.org/10.1002/2015GL063719

Saito, R., Patra, P. K., Sweeney, C., Machida, T., Krol, M., Houweling, S., et al. (2013). TransCom model simulations of methane: Comparison of vertical profiles with aircraft measurements. *Journal of Geophysical Research: Atmospheres*, *118*, 3891–3904. https://doi.org/10.1002/jgrd.50380

Sarin, M., Kumar, A., Srinivas, B., Sudheer, A. K., & Rastogi, N. (2010). Anthropogenic sulphate aerosols and large Cl-deficit in marine atmospheric boundary layer of tropical Bay of Bengal. *Journal of Atmospheric Chemistry*, *66*(1-2), 1–10. https://doi.org/10.1007/s10874-011-9188-z

Schwietzke, S., Sherwood, O. A., Bruhwiler, L. M. P., Miller, J. B., Etiope, G., Dlugokencky, E. J., et al. (2016). Upward revision of global fossil fuel methane emissions based on isotope database. *Nature*, *538*(7623), 88–91. https://doi.org/10.1038/nature19797

Shirai, T., Machida, T., Matsueda, H., Sawa, Y., Niwa, Y., Maksyutov, S., & Higuchi, K. (2012). Relative contribution of transport/surface flux to the seasonal vertical synoptic CO_2 variability in the troposphere over Narita. *Tellus B*, *64*(1). https://doi.org/10.3402/tellusb.v64i0.19138

Shtabkin, Y. A., Moiseenko, K. B., Skorokhod, A. I., Vasileva, A. V., & Heimann, M. (2016). Sources of and variations in tropospheric CO in Central Siberia: Numerical experiments and observations at the Zotino tall tower observatory, *Izvestiya*. *Atmospheric and Oceanic Physics*, *52*(1), 45–56. https://doi.org/10.1134/S0001433816010096

Singh, H. B., Cai, C., Kaduwela, A., Weinheimer, A. J., & Wisthaler, A. (2012). Interactions of fire emissions and urban pollution over California: Ozone formation and air quality simulations. *Atmospheric Environment*, *56*, 45–51. https://doi.org/10.1016/j.atmosenv.2012.03.046

Sklyadneva, T. K., Ivlev, G. A., Belan, B. D., Arshinov, M. Y., & Simonenkov, D. V. (2015). The radiation regime of Tomsk in conditions of a smoky haze. *Atmospheric and Oceanic Optics*, *28*(3), 215–222.

Snider, G., Weagle, C. L., Murdymootoo, K. K., Ring, A., Ritchie, Y., Stone, E., et al. (2016). Variation in global chemical composition of PM 2.5: Emerging results from SPARTAN. *Atmospheric Chemistry and Physics*, *16*(15), 9629–9653. https://doi.org/10.5194/acp-16-9629-2016

Strode, S. A., & Pawson, S. (2013). Detection of carbon monoxide trends in the presence of interannual variability. *Journal of Geophysical Research: Atmospheres*, *118*, 12,257–12,273. https://doi.org/10.1002/2013JD020258

Surawski, N. C., Sullivan, A. L., Roxburgh, S. H., & Polglase, P. J. (2016). Estimates of greenhouse gas and black carbon emissions from a major Australian wildfire with high spatiotemporal resolution. *Journal of Geophysical Research: Atmospheres*, *121*, 9892–9907. https://doi.org/10.1002/2016JD025087

Trentmann, J., Andreae, M. O., & Graf, H.-F. (2003). Chemical processes in a young biomass-burning plume. *Journal of Geophysical Research*, *108*(D22), 4705. https://doi.org/10.1029/2003JD003732

Trentmann, J., Fruh, B., Boucher, O., Trautmann, T., & Andreae, M. O. (2003). Three-dimensional solar radiation effects on the actinic flux field in a biomass-burning plume. *Journal of Geophysical Research*, *108*(D17), 4558. https://doi.org/10.1029/2003JD003422

Van Drooge, B. L., Sicard, M., Stohl, A., Fontal, M., Bravo, N., Munoz, A., et al. (2016). Detection and simulation of wild fi re smoke impacting a Mediterranean urban atmosphere. *Atmospheric Pollution Research*, *7*(3), 494–502. https://doi.org/10.1016/j.apr.2015.12.003

Vasileva, A., & Moiseenko, K. (2013). Methane emissions from 2000 to 2011 wild fi res in Northeast Eurasia estimated with MODIS burned area data. *Atmospheric Environment*, *71*, 115–121. https://doi.org/10.1016/j.atmosenv.2013.02.001

Virkkula, A., Pohja, T., Aalto, P. P., Keronen, P., Schobesberger, S., Clements, C. B., et al. (2014). Airborne measurements of aerosols and carbon dioxide during a prescribed fire experiment at a boreal forest site. *Boreal Environment Research*, *19*(B), 153–181.

Vivchar, A. V., Moiseenko, K. B., & Pankratova, N. V. (2010). Estimates of carbon monoxide emissions from wildfires in northern Eurasia for Airquality assessment and climate modeling, *Izvestiya*. *Atmospheric and Oceanic Physics*, *46*(3), 281–293. https://doi.org/10.1134/S0001433810030023

Wigder, N. L., Jaffe, D. A., & Saketa, F. A. (2013). Ozone and particulate matter enhancements from regional wildfires observed at Mount Bachelor during 2004–2011. *Atmospheric Environment*, *75*, 24–31. https://doi.org/10.1016/j.atmosenv.2013.04.026

Williamson, G. J., Prior, L. D., Jolly, W. M., Cochrane, M. A., Murphy, B. P., & Bowman, D. M. J. S. (2016). Measurement of inter- and intra-annual variability of landscape fire activity at a continental scale: The Australian case. *Environmental Research Letters*, *11*(3), 035003. https://doi.org/10.1088/1748-9326/11/3/035003

Xiong, X., Barnet, C. D., Zhuang, Q., Machida, T., Sweeney, C., & Patra, P. K. (2010). Mid-upper tropospheric methane in the high Northern Hemisphere: Spaceborne observations by AIRS, aircraft measurements, and model simulations. *Journal of Geophysical Research*, *115*, D19309. https://doi.org/10.1029/2009JD013796

Xueref-Remy, I., Bousquet, P., Carouge, C., Rivier, L., & Ciais, P. (2011). Variability and budget of CO2 in Europe: Analysis of the CAATER airborne campaigns—Part 2: Comparison of CO_2 vertical variability and fluxes between observations and a modeling framework. *Atmospheric Chemistry and Physics*, *11*(12), 5673–5684. https://doi.org/10.5194/acp-11-5673-2011

Yang, M. Y. M., Vay, S. A., Stoh, A., Choi, Y., Diskin, G. S., Sachse, G. W., & Blake, D. R. (2012). Chemical composition of tropospheric air masses encountered during high altitude flights (11.5 km) during the 2009 fall Operation Ice Bridge field campaign. *Journal of Geophysical Research*, *117*, D17306. https://doi.org/10.1029/2012JD017858

Zvyagintsev, A. M., Blum, O. B., Glazkova, A. A., Kotel'nikov, S. N., Kuznetsova, I. N., Lapchenko, V. A., et al. (2011). Air pollution over European Russia and Ukraine under the hot summer conditions of 2010, *Izvestiya*. *Atmospheric and Oceanic Physics*, *47*(6), 699–707. https://doi.org/10.1134/S0001433811060168