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On measurements of aerosol particles and greenhouse gases in Siberia and future research needs

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The role of the world's boreal forest for our understanding of the climate system is indisputable. Due to the large area covered, the forest's biophysical (e.g. surface energy balance, albedo) and biogeochemical (e.g. bidirectional exchange of greenhouse gases or aerosol precursors) processes are known to affect today's climate, and will need to be accounted for in studies of climate feedbacks in response to anthropogenic warming. However, observations that are needed to develop and evaluate terrestrial and climate models are still relatively scarce, especially for the Siberian part of the boreal forest. Here, we present a short overview of aerosol and greenhouse gas measurements over Siberia, aiming to also survey a large fraction of the existing literature in Russian. We aim to highlight areas of least data coverage and argue that, due to the importance of Siberia in the global climate system, a coordinated research program is needed to address some of the open research questions: The Pan Siberian Experiment.

Introduction

Siberia covers almost 10% of the Earth's land area and subsumes a vast range of ecosystems, including deciduous and coniferous forests, various types of mires, steppe, tundra, arctic desert, abandoned croplands and cultivated croplands (Fig. 1). A major fraction of the world's boreal forests is located in Siberia, making this area an important source of biogenic volatile organic compounds (Guenther *et al*. 1995, Rinne *et al*. 2009, Timkovsky *et al*. 2010) and natural aerosol particles (e.g. Tunved *et al*. 2006, Dal Maso *et al*. 2008). Western Siberian lowlands constitute the largest mire area in the world, which needs to be taken into account when investigating the global budgets of greenhouse gases (Friborg *et al*. 2003). The high Siberian Arctic land areas are surrounded by ice-covered oceans, and the area itself contains thick permafrost layers with carbon rich soils capable of very high CO_2 or CH_4 emissions if thawed. Discontinuous permafrost areas in Siberia are also very extensive.

The climate is very continental in central

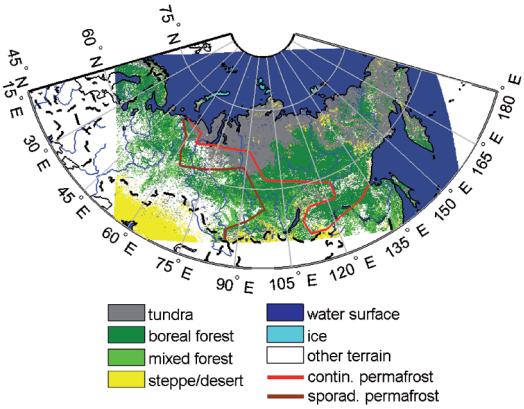

Fig. 1. Ecoregions of Russia and Scandinavia.

Siberia and maritime in its eastern parts. Current and future warming associated with climate change is expected to cause dramatic environmental changes in Siberia (e.g. Diakonov and Romanova 2004, Kotlyakov 2007, Walker 2007, Rinke *et al*. 2008, Tunved *et al*. 2008, Bulygina *et al*. 2009, Swann *et al*. 2010). As part of this process, the budgets of greenhouse gases, as well as the emissions of reactive trace gases like volatile organic compounds and subsequent natural aerosol production, may be significantly affected over this region. Better understanding of these issues requires detailed information on how greenhouse gases, aerosols and aerosol precursor gases are connected with climate via physical, chemical, meteorological and biological processes occurring in the atmosphere and at the atmosphere–biosphere interface.

In spite of its importance in view of impacts of and feedbacks to changing climate, only a relatively small number of investigations on biogeochemical cycles have been performed across the Siberian area to date (Groisman and Soja 2009, Gordov and Vaganov 2010). In what follows we give a short overview of the recent results published in literature that link concentration, fluxes and processes related to aerosols and greenhouse gases (particularly, carbon dioxide and methane) over this region. Based on this overview, we highlight some of the central open science questions related to Siberian ecosystem–climate interactions, including anthropogenic activities, and propose a pan Siberian experiment to address these questions.

Overview of Siberian observations

A large fraction of scientific literature on aerosol and gas research in Siberia has been documented in scientific articles written in Russian. An inquiry into its availability revealed that only a part of this literature is accessible on the Internet. Nevertheless, this literature, both in English and in Russian, is deemed sufficient for constructing a consistent image of scientific research conducted in Siberia.

Despite the large geographical and climatological expanse when considering the whole Siberia as a study area, the conducted research did not have a uniform geographic distribution, and was conducted mainly in the western Siberia (Fig. 2). There are several long-term measurement stations, towers and masts equipped with diverse instrumentation and providing information on multiple gas and particle parameters (Table 1); some of these are stations in Tomsk, Plotnikovo, Mukhrino and Vasyuganye. Various measurement campaigns utilizing both stationary and mobile platforms in most regions of Siberia have also contributed to increasing the spatial coverage (Tables 1 and 2). Measurements in Siberia have been conducted using both airborne and ground-based vehicles. Some of the measurements were conducted in remote areas (Zyryanov *et al*. 2008), but also areas subject to major anthropogenic influence were investigated (e.g. Arshinov *et al*. 2006b, Inisheva *et al*. 2007). It should be noted that while greenhouse gas measurements have mainly been conducted in background and remote areas, studies of aerosol and related gases have concentrated on urban centers. Measurement campaigns generally provided short-term data (few weeks to few years), with permanent stations being the main sources of long-term information. Gas and particle data collection has been carried out in Siberia since the early 1990s.

Aerosol measurements

Aerosol physical, chemical and optical properties

Aerosol number size distributions have been

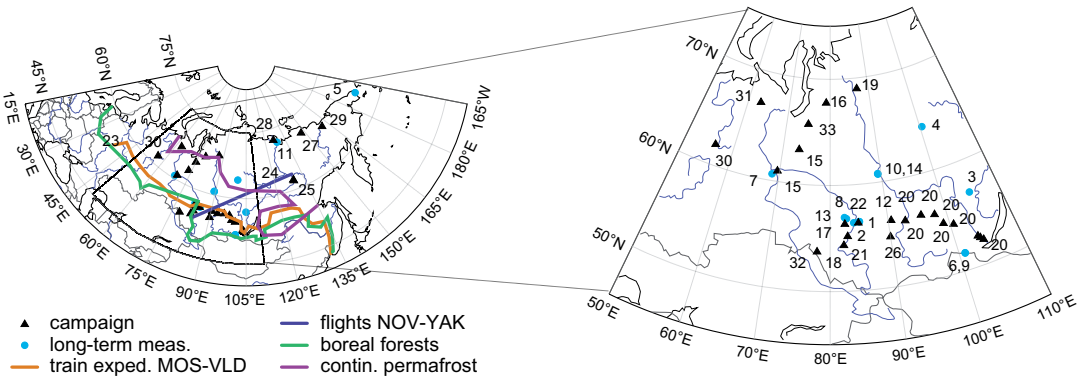

Fig. 2. Measurement sites and campaigns in Siberia. For numbers and names of the sites *see* Table 1..

measured in Siberia mainly with diffusion aerosol spectrometers (e.g. Julanov *et al*. 2002), differential mobility particle sizers and various optical counters. The first two types of instruments typically performed measurements of sizes of a few nm of particle mobility diameter and extended up to sizes of a few hundred of nm (Table 3). The applied optical counters covered typically the particle diameter range from a few hundred nm to a few micrometers (Table 3).

Siberian aerosol number size distributions display a modal structure, with one to three modes present between about 25 and a few hundred nm, and frequently a nucleation mode below 25 nm (Koutsenogii 1997, Arshinov and Belan 2000, Vartiainen *et al*. 2007, Heintzenberg *et al*. 2011). Some investigators also measured supermicron particles (Koutsenogii 1997), but there is very little information on the shape of the aerosol size distribution above a few hundred nm. The traditional Aitken (25–100 nm) and accumulation (100–1000 nm) modes can usually be separated at remote sites, but not necessarily at sites affected by local or nearby pollution sources. The average particle number size distributions tend to peak close to the diameter of 100 nm, even though the exact shape of the number size distribution varies between the individual cases (Vartiainen *et al*. 2007, Heintzenberg *et al*. 2011). At practically all sites, aerosol number concentrations seem to be dominated by particles smaller than 100 nm in diameter.

The average total particle number concentrations were observed to range from less than 1000 cm^{-3} to about 5000 cm^{-3} in remote Siberian sites (Dal Maso *et al*. 2008, Heintzenberg *et al*. 2011). At a given site, particle number concentrations vary by 1–2 orders of magnitude (5 to 95 percentile values), depending on the origin of measured air masses, season and the time of day. Overall, aerosol number concentrations at remote Siberian sites are comparable to those over the Scandinavian boreal forest (Dal Maso *et al*. 2007), but clearly higher than concentrations typical for remote marine areas or cleanest continental locations (Heintzenberg *et al*. 2000, Andreae 2007). At Siberian locations affected by pollution, total number concentrations were found to be larger, typically in the range of $1–5 \times 10^4$ cm^{-3} (Vartiainen *et al*. 2007).

Compared with the data on aerosol number size distributions, much less information is available on the mass concentrations or chemical composition of Siberian aerosols. Kuokka *et al*. (2007) used a moving laboratory placed in a train traversing Siberia for chemical aerosol measurements. They found that the aerosol $PM_{2.5}$ mass concentrations varied between about 4 and 35 μg m^{-3} with an average of 22 μg m^{-3}. The aerosol chemical composition was dominated by carbonaceous matter and sulfate, with minor components being ammonium, nitrate, some other inorganic ions and many trace elements. The presence of sulfate and nitrate in Siberian aerosols has been reported in a few other investigations as well, along with indications of crustal material, sea salt and polycyclic aromatic hydrocarbons (Koutsenogii *et al*. 1996, 1998, Van Malderen *et al*. 1996, Koutsenogii 1997, Tolmachev 1999, Arshinov *et al*. 2002, Belan *et*

Table 1. Details of the selected measurement sites and campaigns in Siberia and the Russian Far East (*see* Fig, 1 for locations on a map and Table 2 for references) (* = not depicted on the map).

Site	Name, type	Location	Measured parameters	Time interval
1	Base Experimental Complex (BEC) station, suburban	Tomsk, 56°29´N, 85°04´E, 170 m a.m.s.l.	aerosol, gas and meteo data	n/a
2	"Fonovyi" station, background	56°25´N, 84°04´E, 80 m a.m.s.l.	aerosol, gas and meteo data	from 2006 onwards
3	Listvyanka station, background, lakeshore	56°54´N, 104°54´E	aerosol data	from 1990 onwards
4	mast, background	64°12´N, 100°27´E	CO_2, heat (EC), meteo data	from 1990 onwards
5	mast, background	65°35´N, 171°00´W	CO_2, meteo data, radiation	from 2000 onwards
6	Mondy station, background	51°39´N, 100°55´E, 2005 m a.m.s.l.	aerosol, gas and meteo data	from 1996 onwards
7	Mukhrino station, background	60°38´N, 68°37´E	CO_2, CH_4, meteo data, radiation	from 2007 onwards
8	Plotnikovo station, suburban, swamp	56°50´N, 82°51´E	CH_4, soil data	from 1993 onwards
9	Sayan Solar Observatory, background, mountainous	51°37´N, 100°55´E, 2005 m a.m.s.l.	aerosol and meteo data	n/a
10	station, background	60°45´N, 89°23´E	CO_2, meteo data, trunk respiration, profile measurements	from 1998 onwards
11	Tiksi observatory, arctic	71°40´N, 128°40´E	aerosol, gas and meteo data	meteo from 1930s onwards, gas and aerosol from 2010 onwards
12	TOR station, urban	Tomsk, 56°29´N, 85°03´E	aerosol, gas and meteo data, surface albedo	from 1992 onwards
13	Vasyuganye station, background	56°57´N, 82°30´E	CO_2	from 1996 onwards
14	ZOTTO tower, background, boreal	60°48´N, 89°21´E	aerosol and meteo data	from 2006 onwards
15	campaign, floodplain	63°15´N, 73°26´E and 60°59´N, 69°34´E	CH_4 flux	August 2007
16	campaign	67°46´N, 79°02´E	CH_4 flux, soil data	August and December 2009
17	campaign	56°03´–56°37´N, 82°22´–82°42´N	CO_2 flux, meteo data	1998–1999
18	campaign (aircraft)	Novosibirsk (54°08´–54°33´N, 81°51´–82°40´N)	aerosol and gas data	1999–2008 (monthly)
19	campaign (truck and aircraft)	Norilsk (68°25´–71°00´N, 84°00´–90°00´N)	aerosol, gas and meteo data	November 2002 and August 2004
20	campaign (truck)	Irkutsk, Angarsk, Usol'ye-Sibirskoe, Tulun, Nizhneudinsk, Taishet, Kansk, Krasnoyarsk, Achinsk	aerosol, gas and meteo data, radiation	February 2004
21	campaign (truck)	Novosibirsk, 55°1´N, 82°56´E	aerosol, gas and meteo data	March and July 2004
22	campaign, peat deposit "Tagan"	56°23´N, 84°52´E	carbonaceous greenhouse gases	1996–2001
23	TROICA-9 expedition, campaign (train)	Moscow to Vladivostok	aerosol, gas and meteo data	October 2005

24	YAK-AEROSIB, campaign (aircraft)	Novosibirsk ↔ Yakutsk	aerosol and gas data	April and September 2006
25	campaign, larch forest	62°15´N, 129°37´E, 220 m a.m.s.l.	CO_2 flux	July–November 2000, April–September 2001
26	campaign, steppe	54°45´N, 90°00´E	CO_2 flux	May–September 2004
27	campaign, tundra	70°50´N, 147°30´E, 10 m a.m.s.l.	CO_2, CH_4 flux	2003–2006, summer
28	campaign, tundra	72°22´N, 126°30´E	CO_2, CH_4 flux	July–October 2003, May–July 2004
29	campaign, tundra	68°37´N, 161°20´E	CO_2, CH_4 flux	2002–2005, summer
30	campaign, mire	61°56´N, 56°13´E	CH_4 flux	April–May 2008
31	campaign, tundra wetland	67°23´N, 63°22´E	CO_2, CH_4 flux	June–September 1999, March 2000, June–September 2001
32	campaign, steppe	53°44´N 77°54´E	aerosol data	February–March, August–September 1992
33	campaign, tundra	65°52´N, 74°58´E	CO_2, CH_4 flux	July, August 2005
34*	campaign (aircraft)	Tomsk (approx. 56°30´N, 86°00´E)	aerosol data	1986–1988
35*	campaign (aircraft)	78°N, 85°E; 81°N, 124°E; 60°N, 88°E; 69°N, 87°E; 74°N, 65°E	aerosol data	summer 1993

al. 2001, 2005b, Yermakov *et al*. 2007). While probably a major component of the aerosol, to our knowledge no study published so far has reported direct measurements of organic particulate matter in this region.

Aerosol optical properties have been reported in a few studies. Koutsenogii (1997) measured the aerosol scattering coefficient in the Novosibirsk region and found higher averaged values in winter than in summer, congruent with near-surface observations made during aircraft campaigns by Panchenko *et al*. (1996) and Kozlov *et al*. (2009). A similar seasonal cycle with a summer minimum and winter maximum was reported for the aerosol absorption coefficient by Heinzenberg *et al*. (2011) on the ZOTTO (Zotino Tall Tower Facility) tower in central Siberia. Long-term measurements of the aerosol optical depth (AOD) over four Siberian cities was reported by Nikolashkin *et al*. (2009). The AOD was observed to be highest in spring or summer and to decrease gradually toward winter. Similar seasonal variation of AOD, as estimated based on the Russian actinometric network in the years 1976–2003, was reported by Plakhina *et al*. (2007), although they also stated that this trend vanished in regions of low AOD. The same study estimated that, in general, AOD shows a gradually decreasing trend in Russia in the west to east direction.

Aerosol spatial and temporal variability and sources

Based on aircraft measurements, total aerosol number concentrations have usually been observed to decrease gradually with increasing height over Siberia (Arshinov and Belan 2004, Kozlov *et al*. 2009, Paris *et al*. 2009). Some of the measured profiles showed, however, elevated particle number concentrations in the mid to upper troposphere, especially for particles smaller than 100 nm in diameter. Long-term measurements at the ZOTTO facility showed slightly higher concentrations of all integral aerosol parameters at the 50 m height compared with the 300 m height (Heintzenberg *et al*. 2011). In addition, aerosol scattering and absorption coefficient profiles, based on aircraft studies, show a generally decreasing trend with altitude, but also

suggest some seasonal differences (Panchenko *et al*. 1996, Kozlov *et al*. 2009). The campaign-type measurements in summer with airborne lidar show a spatial decrease in surface aerosol scattering coefficient towards the north, while the scattering at higher altitudes is deduced to largely depend on the atmospheric stratification (Khattatov *et al*. 1997). No clear picture about the latitudinal or longitudinal distribution of aerosol concentration over Siberia can be obtained based on available measurements.

Table 2. References for corresponding stations and campaigns in Table 1 (* = not depicted on the map).

Site	Reference
1	Arshinov *et al*. 2007
2	Arshinov *et al*. 2007
3	Dal Maso *et al*. 2008
4	Zyryanov *et al*. 2008
5	Zamolodchikov *et al*. 2005
6	Yermakov *et al*. 2007
7	Glagolev *et al*. 2010a
8	Glagolev and Smagin 2006, Arshinov *et al*. 2006f, Glagolev *et al*. 2010a
9	Golobokova *et al*. 2006
10	Vaganov *et al*. 2005
11	Asmi *et al*. 2010
12	Arshinov *et al*. 1994, Arshinov *et al*. 1999, Arshinov *et al*. 2006bf, Dal Maso *et al*. 2008
13	Golovatskaya and Dyukarev 2007
14	Heinzenberg *et al*. 2011
15	Glagolev and Suvorov 2007
16	Glagolev *et al*. 2010a
17	Inisheva 2005
18	Arshinov and Belan 2004, Kozlov *et al*. 2009, Paris *et al*. 2009
19	Arshinov *et al*. 2006cde, Arshinova *et al*. 2006
20	Belan *et al*. 2005a
21	Belan *et al*. 2005b
22	Inisheva *et al*. 2007
23	Kuokka *et al*. 2007, Vartiainen *et al*. 2007
24	Paris *et al*. 2009
25	Dolman *et al*. 2004
26	Belelli Marchesini *et al*. 2007
27	van Huissteden *et al*. 2005, van der Molen *et al*. 2007
28	Kutzbach *et al*. 2007, Wille *et al*. 2008
29	Corradi *et al*. 2005, Merbold *et al*. 2009
30	Gažovič *et al*. 2010
31	Heikkinen *et al*. 2002, Heikkinen *et al*. 2004
32	van Malderen *et al*. 1996
33	Naumov *et al*. 2007
34*	Panchenko *et al*. 1996
35*	Khattatov *et al*. 1997

Total aerosol number concentrations have usually been observed to be higher in summer than in winter (Heintzenberg *et al*. 2011), even though seasonal changes in particle number concentrations may be different between the boundary layer and the free troposphere (Paris *et al*. 2009). Contrary to this, quantities proportional to the aerosol mass or volume concentrations have been found to display a winter maximum (Panchenko *et al*. 1996, Koutsenogii, 1997, Kozlov *et al*. 2009, Heintzenberg *et al*. 2011). Besides the seasonal variability, some studies have reported a clear diurnal cycle for aerosol number concentrations, with typically a daytime maximum (Arshinov and Belan 2000, Dal Maso *et al*. 2008). All measurement time series are too short to deduce any long-term trend in total aerosol number concentrations. However, a few trend estimates are available for particles larger than about 400 nm in diameter, as well as for some chemical compounds (Belan and Tolmachev 1996, Arshinov *et al*. 2002).

Differences in the temporal variability between the aerosol number concentrations and mass-related quantities are indicative of the variability of the sources, or sinks, affecting particles of different sizes. Identified natural aerosol sources in Siberia include mineral dust and sea salt (Tolmachev 1999, Belan *et al*. 2001), primary biogenic particles (Matthias-Maser *et al*. 2000, Borodulin *et al*. 2005), and atmospheric new particle formation discussed in more detail in the next subsection.

An important anthropogenic source of Siberian aerosols, identified in several studies, is biomass burning (Soja *et al*. 2004, Kuokka *et al*. 2007, Kozlov *et al*. 2008, Vivchar *et al*. 2010, Heintzenberg *et al*. 2011). Siberian biomass burning is of particular international interest due to its suggested significant impacts on the arctic climate (e.g. Stohl 2006, Warneke *et al*. 2010). Yet, the estimates of the source strength of particulate matter and its chemical content by this burning present a large spread of values (Conard *et al*. 2002, Soja *et al*. 2004, Samsonov *et al*. 2005, McRae *et al*. 2006, Kozlov *et al*. 2008). A major fraction of this biomass burning comes from frequent forest fires and agricultural fires taking place in spring and summer (e.g. Sukhinin *et al*. 2004, Korontzi *et al*. 2006),

Table 3. Gas and aerosol instrumentation at three selected stations (refer to Table 1).

Parameter	Unit of measurement	Instrument	Range	Error (%)	Comments	Location
CO_2	%	L-061-2	0.0001–0.2	20	differential absorption in the IR	TOR-Station
CO_2	ppm	L-061-02 (GMM222)	0–10000	20		"Fonovyi", BEC
CO	mg m^{-3}	L-061-2	0.1–200	20	electrochemical sensor	TOR-Station
CO	mg m^{-3}	K-100	0–400	20		"Fonovyi", BEC
O_3	μg m^{-3}	3-02P	0–1000	15	chemiluminescence	TOR-Station, "Fonovyi", BEC
NO_2	μg m^{-3}	P-310-11 and P310-A	0–1000	25	chemiluminescence	TOR-Station, "Fonovyi", BEC
NO	μg m^{-3}	P-310-11 and P310-A	0–1000	25	chemiluminescence	TOR-Station, "Fonovyi", BEC
H_2S	μg m^{-3}	SV-320	0–200	15		BEC
NH_3	μg m^{-3}	N-320	0–1000	15		BEC
SO_2	μg m^{-3}	C310	0–2000	25		"Fonovyi", BEC
$N(r)$	cm^{-3}	photoelectrical particle counter A3-5	1–300	20	0.2 μm < $r \leq$ 10 μm; 12 channels in the 0.2 μm < $r \leq$ 5 μm range	TOR-Station
$N(r)$	cm^{-3}	diffusional aerosol spectrometer DAS	1–10000	20	0.003 μm $\leq r \leq$ 0.2 μm; 15 channels in the 0.008 μm $\leq r \leq$ 0.2 μm range	TOR-Station, BEC
N	cm^{-3}	Aerosolmeter, GRIMM 1108	0–300	25		"Fonovyi", BEC
α (0.55 μm)	km^{-1}	photoelectrical nephelometer FAN	0.001–1	7		TOR-Station

whereas in winter domestic heating may be important. Indicative of anthropogenic combustion sources, elevated number concentrations of Aitken and accumulation mode particles have been observed to correlate with gaseous nitrogen oxide, carbon monoxide and sulfur dioxide concentrations in Siberia (Vartiainen *et al*. 2007). Moreover, some studies reported the evidence of long-range transported anthropogenic particles of Siberian aerosols with suggested origin in industrial regions of north Kazakstan and former Soviet Union (Van Malderen *et al*. 1996, Koutsenogii *et al*. 1998). Van Malderen *et al*. (1996) stated that these particles have potential to be further transported and deposited in the Arctic, which was also supported by the recent modeling studies (e.g. Stohl 2006). Similarly, analyses of air mass transport patterns have shown that the lowest particle number and mass concentration are usually observed in the Arctic air masses originating from northern Siberia or the Arctic Ocean, whereas most polluted air masses can be traced to industrial regions in Russia, Kazakstan and even China (Kuokka *et al*. 2007, Heintzenberg *et al*. 2011).

New-particle formation over Siberia

Atmospheric new-particle formation taking place in Siberia is of high interest because the region encompasses a major fraction of the whole boreal forest area. Studies conducted in the Scandinavian part of the area suggest that new-particle formation associated with boreal forest emissions is a frequent phenomenon, being the dominant source of the particles in terms of their number concentration during the summer part of the year (Tunved *et al*. 2006, Dal Maso *et al*. 2007). If the same applies to the whole boreal forest zone, natural emissions from boreal forests might induce regionally significant indirect radiative effects (Kurtén *et al*. 2003, Spracklen *et al*. 2008, Tunved *et al*. 2008, Lihavainen *et al*. 2009).

While indications of atmospheric new-particle formation have been reported in several studies conducted in Siberia (Bashurova *et al*. 1992, Koutsenogii and Jaenicke 1994, Arshinov and Belan 2000, Vartiainen *et al*. 2007, Heintzenberg *et al*. 2011), including the free troposphere (Arshinov and Belan 2004, Paris *et al*. 2009), systematic investigations of this phenomenon are very limited in this region. Dal Maso *et al*. (2008) analysed year-long measurement data sets from two Siberian locations and found that new-particle formation events took place in roughly 10% of the days. Such event frequency is clearly lower than that observed over Scandinavian boreal forests (Dal Maso *et al*. 2007, Kristensson *et al*. 2008) or in most other European sites with long-term measurements (Manninen *et al*. 2010). Dal Maso *et al*. (2008) reported that new-particle formation events are most common in the spring, but it must be noted that the available measurement data are insufficient for drawing firm conclusions about the seasonal behavior of Siberian new-particle formation.

Consistent with measurements made elsewhere within continental boundary layers (Kulmala *et al*. 2004, Kerminen *et al*. 2010), new-particle formation over Siberia seems to be photochemically driven (Arshinov and Belan 2000, Arshinov *et al*. 2006a, Dal Maso *et al*. 2008). The formation rates of new particles have been estimated in a few studies as well. Earlier studies reported relatively low new-particle formation rates (Arshinov and Belan 2000, 2004), whereas later investigations obtained rates on the order of 0.4–0.5 cm^{-3} s^{-1} (Arshinov *et al*. 2006a, Dal Maso *et al*. 2008). The same studies estimated the growth rates of a few nm $hour^{-1}$ for newly-formed particles. Both the new-particle formation and growth rates observed in Siberia are comparable to those found in Scandinavian boreal forests (Dal Maso *et al*. 2007, Kristensson *et al*. 2008). This indicates that atmospheric new-particle formation taking place over Siberia has clearly potential to influence the regional aerosol population and, thereby, the whole climate system.

Greenhouse gas measurements

Greenhouse gas (here, mainly carbon dioxide and methane) measurements have been conducted over Siberia at different levels of temporal and spatial integration. The smallest of the spatial scales addressed, a few dm^2, are studied

via flux measurements using chambers (Christensen *et al*. 1998, Zamolodchikov and Karelin 2001). This method was used, for example, to study the effect of temperature, water table depth and plant community variations on CO_2 and CH_4 exchange in boreal mires (Naumov *et al*. 1994, Shibistova *et al*. 2002, Riutta *et al*. 2007). The micrometeorological eddy covariance method is a nonintrusive way of measuring the net gas balance at the ecosystem level, on an area typically including several hectares (Baldocchi 2003). In case of CO_2, flux measurements can in principle be applied as a year-round operation since the sensors are sufficiently robust to allow for the necessary automation. By using a chamber technique on leaves and branches, it is straightforward to measure net photosynthesis rates during the day and total respiration ($Resp_{tot}$) by darkening the chamber, or by measuring at night. Chambers used to enclose soil and understory measure during the day the net exchange of carbon, which is the sum of Gross Primary Production (GPP) and $Resp_{tot}$, and solely $Resp_{tot}$ at night (which is the sum of autotrophic and heterotrophic respiration). Similarly, on an ecosystem scale integrative measurement techniques, such as micrometeorologial method, observe the net flux between GPP and $Resp_{tot}$, called Net Ecosystem Exchange (NEE), directly integrated over few hectares. If one is interested in the component fluxes GPP and $Resp_{tot}$, they have to be estimated indirectly (Reichstein *et al*. 2005). If one uses a chamber method and would like to know NEE, simple response modeling and temporal extrapolation are usually involved. Furthermore, if spatially averaged information, for example to ecosystem scale, is estimated from chamber data, spatial extrapolation methods are needed. Long-term eddy-covariance sites have been organized globally under FLUXNET (Baldocchi *et al*. 2001), which has regional networks such as EUROFLUX (Reichstein *et al*. 2007) and ASIAFLUX (Kato and Tang 2008).

The Global Atmosphere Watch (GAW) programme of the World Meteorological Organization (WMO) coordinates observations of atmospheric trace gas concentrations at a global network of stations (http://gaw.empa.ch/gawsis/). GAW aims at providing reliable scientific data on the chemical composition of the atmosphere and its natural and anthropogenic change focusing on global networks for GHGs, ozone, UV, aerosols, selected reactive gases, and precipitation chemistry. From Russia, CO_2 and CH_4 concentrations from flask samples collected in Teriberka, Kola Peninsula, have been reported to the GAW database (http://gaw.kishou.go.jp/wdcgg/) (Paramonova *et al*. 2001). The GAW main site nearest to Russia is at Pallas, in northern Finland (Aalto *et al*. 2007), which has been in operation since 1996. Recently, high-quality GHG monitoring was conducted at a tall tower site in central Siberia (Kozlova *et al*. 2008). The most utilized global standardized data source is the co-operative flask sampling network maintained by the U.S. National Oceanic & Atmospheric Administration (NOAA) Earth System Research Laboratory (ESRL) Carbon Cycle Greenhouse Gases (CCGG) group (http://www.esrl.noaa.gov/gmd/ccgg/index.html), which aims to document the spatial and temporal distributions of carbon-cycle gases and provide essential constraints to our understanding of the global carbon cycle. Air samples are collected approximately weekly from a globally distributed network of sites and analyzed in Boulder by CCGG. From Russia, air samples collected in Obninsk, 100 km SW from Moscow, are analyzed within this network. In 2011 (or 2012), the NOAA co-operative flask sampling network is starting a new site in Tiksi on the coast of the Laptev Sea. These data give area-averaged information about daily, seasonal and annually changing atmospheric burdens, but cannot be used to infer exchange fluxes.

On the landscape level, spatial variations of greenhouse gas concentrations have been measured by aircraft (Paris *et al*. 2008). Aircraft platforms can also be used to infer vertical GHG fluxes, by equipping the aircraft with fast response turbulence instrumentation (Desjardins *et al*. 1994). Otherwise, landscape level uptake and emissions have to be calculated from concentration profiles combined by budgeting methods such as the CBL method (Lloyd *et al*. 2001), or by calculating land–atmosphere fluxes from a network of vertical concentration profiles analyzed together with wind fields (Crevoisier *et al*. 2010). Vertical profiles have been used also to verify and improve large-scale inversion model results (Stephens *et al*. 2007). Data from aircraft plat-

forms have the advantage of spatial integration, but become expensive especially if used for long-term monitoring. The total column GHG burden has also been measured by ground-based Fourier Transform Spectrometers using detailed spectral information on absorbed solar radiation (Geibel *et al*. 2010). The column observations will be used in regional and global carbon cycle models and satellite validation. Globally this network is organized as Total Column Carbon Observation Network (http://www.tccon.caltech.edu/).

In summary, flux measurements usually are relatively small in terms of their spatial coverage from soil or leaf chambers to flux towers. Based on these types of measurements, annual balances have been estimated by extrapolating observations at a number of specific ecosystems to country scale using land use information (for methane, *see* e.g. Glagolev *et al*. 2010b). Carbon storages and fluxes have been estimated from biomass and soil carbon inventories (e.g. Kurganova *et al*. 2010). Top-down methods use atmospheric concentration data and dispersion models to estimate emissions. Reshetnikov *et al*. (2009) used this method to estimate methane emissions from western Siberian gas fields. Large scale emission fields have been produced from atmospheric concentration data by inversion models, which optimize *a-priori* flux intensity maps using atmospheric gas concentrations observations to reach optimized spatial flux intensity fields (Gurney *et al*. 2002). The large-scale top-down estimates were used to develop bottom-up carbon accounting estimates and their uncertainties (Gusti and Jonas 2010).

Soil and leaf enclosure measurements of CO_2

The traditional way of making gas exchange measurements is the chamber method, which has been used in many studies in the western Siberian lowlands. The measurements of carbonaceous greenhouse gases in west Siberia were started in early 1990s in the Tomsk district, in the area where the Great Vasyugan Bog is located, the largest peatland system in the world (53 000 km^2). From the beginning, the station near the Plotnikovo village at the Bakcharsky mire has been the main measurement site (Naumov *et al*. 1994, Panikov *et al*. 1995). The CO_2 emission rates in Vasyugan peatland were investigated by Inisheva and Golovatskaya (2002). During the growing season, highest variations of the CO_2 emissions were in high bog forest and lowest in the open sedge–spaghnum site. Cumulative emissions of CO_2 throughout the vegetative period of 1999 were 80 g(C) m^{-2} at low and high forest bog areas in the lower rim of the bog. In the raised center of the bog, cumulative CO_2 emissions were 60 and 66 g(C) m^{-2} in the sparsely forested and open sedge–spaghnum microsites, respectively. Golovatskaya *et al*. (2008) found positive correlation between temperature and negative correlation between water table height and CO_2 emissions. Golovatskaya and Dyukarev (2009) presented the results of a long-term (1999–2007) investigation of vegetation productivity and CO_2 emissions from the surface of an oligotrophic mire. The studied ecosystems include pine–shrub–sphagnum (PSS) community, a similar community with a low tree stand (LPSS), and sedge–sphagnum fen (SSF). Field measurements of NPP and CO_2 emissions in combination with reference data on methane emissions, winter CO_2 and CH_4 emissions and carbon export by river run-off were used to develop an overall carbon budget of the mire ecosystems. The studied mire ecosystems were found to be net sinks for atmospheric carbon and to accumulate peat. Rates of the actual modern annual carbon accumulation were equal to 21, 112 and 102 g(C) m^{-2} for PSS, LPSS and SSF communities, respectively.

Studies in the central part of the western Siberia, the Khanty-Mansiysk district, have been recently centered around the Mukhrino research station. Summer average CO_2 soil respiration ranged between 5.6 to 11 g(CO_2) m^{-2} d^{-1} at different mire microsites in fen and pine bog ecosystems (Naumov *et al*. 2007). In the northern part of the western Siberia, forest measurements in the tundra subzone with a discontinuous permafrost layer near Pangody showed that summer average CO_2 emissions were 5.4 and 4.6 g(CO_2) m^{-2} d^{-1} in frozen palsa and oligotrophic hollow, respectively (Naumov *et al*. 2007).

A large number of GPP and $Resp_{tot}$ measurements were conducted by transparent and opaque chambers in several locations spanning

the Siberian Arctic region (Zamolodchikov and Karelin 2001). GPP and $Resp_{tot}$ were modelled using biomass and environmental data. Together with landuse and environmental information from Russian tundra, total carbon balances were calculated. The annual GPP of Russian tundra zone for the area of 2350 000 km^2 was estimated at –485.8 Tg(C) and $Resp_{tot}$ at 474.2 Tg(C), resulting in a small net sink of –11.6 Tg(C) (Zamolodchikov and Karelin 2001).

Micrometeorological CO_2 and CH_4 flux measurements

The state-of-the-art method for obtaining the understanding of ecosystem CO_2 or CH_4 exchanges is the eddy covariance technique (Baldocchi *et al*. 1988, Aubinet *et al*. 2000). It is a non-destructive micrometeorological technique that does not affect environmental conditions such as light distribution, temperature, CO_2 and H_2O concentrations and turbulence. Based on eddy flux measurements, various aspects of gas exchange related to e.g. photosynthesis, respiration, soil processes, seasonal cycles and measurement methods have been studied. In this review, we try to present those studies which aim at seasonal or annual balances of CO_2 and CH_4 exchange.

Within the EuroSiberian Carbonflux Project intensive measurements were conducted in central Siberia close to Zotino, as well as in European Russia forest and mire ecosystems. These sites lie outside the permafrost region. Lloyd *et al*. (2002) presented a study of the old pine forest on sandy soil in Zotino (*see* also Valentini *et al*. 2000 for summer measurements in the same region). The forest was a substantial annual sink for CO_2, around 160 g(C) m^{-2}. Flux measurements in three forests, a deciduous birch-dominated, a coniferous spruce-dominated and a mixed forest growing on Cambisol soils near Zotino, were described by Röser *et al*. (2002). Measured uptake for the growing season was 250 g(C) m^{-2} in the *Betula* stand, 100 g(C) m^{-2} in the mixed boreal forest, and 270 g(C) m^{-2} in the *Abies* stand. Interestingly, there was an upland spruce site and another one located on organic soil (60 cm peat layer) where the water-table level was often close to the surface. The upland spruce site was an annual sink of carbon from the atmosphere (144 g(C) m^{-2}), while the organic soil site was a source of carbon (200 g(C) m^{-2}).

Measurements of ecosystem CO_2 and H_2O fluxes and associated climatic variables were conducted over three growing seasons at two bogs: Fyodorovskoye located in European Russia, and Zotino in central Siberia (Arneth *et al*. 2002). In Zotino, the inter-annual variability in climate, and also in the CO_2 balance during the snow-free period, was small. Arneth *et al*. (2002) estimated that the annual sink during two years in Zotino bog was 22 and 36 g(C) m^{-2}. At Fyodorovskoye, summer climate was more variable. During the year which was below-average dry, the mire was a source of CO_2 to the atmosphere, roughly 50 g(C) m^{-2}. During a climatologically normal year, the bog was a sink of CO_2.

The first micrometeorological CO_2 flux measurements in the permafrost region of Sakha (Yakutia) were done in 1993 (Hollinger *et al*. 1995, Arneth *et al*. 1996, Kelliher *et al*. 1997). In the same area, Dolman *et al*. (2004) measured CO_2 fluxes at a Siberian larch (*Larix cajanderii*) site. They estimated that annual carbon sink was 160 g(C) m^{-2} in 2001. Zyryanov *et al*. (2008) studied effects of microclimate on CO_2 exchange at a larch (*Larix gmellini*) site in Evenkia. They estimated that the forest was a carbon sink during the growing season.

Belelli Marchesini *et al*. (2007) measured CO_2 fluxes of natural graminoid small-tussock steppe in the Iyus-Shira region of the Republic of Hakasia. According to micrometeorological measurements, a sink of carbon of 150 g(C) m^{-2} accumulated during the growing season from May to September.

The landscape at tundra locations often consists of microsites of bare soil, vegetated dryer areas and moist vegetated depressions, where methane production may occur in organic soil.

Carbon dioxide and methane fluxes were measured at a tundra site near Chokurdakh, in the lowlands of the Indigirka river in north-east Siberia (van der Molen *et al*. 2007). As compared with those at other tundra sites, net carbon dioxide fluxes were high having an annual sink of 92 g(C) m^{-2}.

Kutzbach *et al*. (2007) investigated the flux of CO_2 between wet arctic polygonal tundra and

the atmosphere by the micrometeorological eddy covariance method in the centre of the Lena River Delta in northern Siberia. They observed a substantial CO_2 sink of 32 g(C) m^{-2} over the summer and an estimated annual net ecosystem CO_2 sink of 19 g(C) m^{-2}.

Carbon dioxide, energy flux measurements and methane chamber measurements were carried out in an Arctic wet tussock grassland located on a flood plane of the Kolyma river in NE Siberia over a summer period of 155 days (Corradi *et al*. 2005). The cumulative annual net carbon flux from the atmosphere to the terrestrial surface was estimated to be about 38 g(C) m^{-2}. During the next year the site was a small source of CO_2 to the atmosphere (Merbold *et al*. 2009). The site was artificially drained by lowering the water table depth by 5–30 cm during the growing season. This resulted in a growing season CO_2 balance of close to zero (Merbold *et al*. 2009).

In Tiksi, close to the Laptev Sea in northern Sakha, we started measuring CO_2 fluxes in summer 2010 on tundra surface where there are moist methane-emitting depressions present (Laurila *et al*. 2010).

Micrometeorological fluxes of CO_2 were measured in late July–September in 2000–2002 in the vicinity of the settlement of Lavrentiya, Chukchi Peninsula (Zamolodchikov *et al*. 2003). The tundra ecosystem under examination was characterized by the well-pronounced cryogenic microrelief, represented by the hummocks and small depressions between them. The dominating vegetation was an assortment of shrubs, weeds and mosses. During 23 July–30 September, the tundra was a carbon sink of 26, 42, 47 g(C) m^{-2} in the years of 2000, 2001 and 2002, respectively (Zamolodchikov *et al*. 2005). Mean air temperature explained the interannual variation.

Wetland methane emissions

In anaerobic soils, methane is produced by the degradation of organic matter. The emission factors have been traditionally measured by chambers. Similar to CO_2, micrometeorological methods are a powerful tool to make seasonal methane emission measurements, which are emerging from Russia as well. Sachs *et al*. (2010) compared chamber and micrometeorological measurements in the wet polygonal tundra in the Lena river delta. They reported that methane emissions responses to the main environmental parameters, water table depth and near-surface turbulence, depended critically on the measurement method. They call for nonintrusive methods to obtain unbiased results.

Wille *et al*. (2008) reported extended growing season measurements from the Lena river delta site. The surface is characterized by wet polygonal tundra, with a micro-relief consisting of raised, moderately dry sites, depressed wet sites, polygonal ponds, and lakes. The annual CH_4 emission measured by the micrometeorological method was estimated to be 2.4 g(C) m^{-2}.

In addition to CO_2 fluxes by micrometeorological technique, CH_4 emissions were measured by the chamber method in the Indigirka lowlands in Sakha Republic (van Huissteden *et al*. 2005). Methane fluxes from arctic tundra soils on a river terrace and floodplain in northeastern Siberia showed a high spatial variability. The CH_4 fluxes on the river terrace compare well with fluxes reported in other studies on tundra CH_4 fluxes. The average CH_4 flux was 4.3 mg m^{-2} h^{-1}, and the average flux for wet microsites was 7.2 mg m^{-2} h^{-1} and for dry microsites 0.18 mg m^{-2} h^{-1}, with negative fluxes occurring locally. The fluxes from floodplain sites were considerably higher, with an average flux of 12.5 mg m^{-2} h^{-1}, an average for wet microsites of 23.4 mg m^{-2} h^{-1}, and an average for dry microsites of 1.6 mg m^{-2} h^{-1}. On average, annual methane emission was estimated at 3.1 g(C) m^{-2} (van der Molen *et al*. 2007).

Merbold *et al*. (2009) reported multiannual emissions from the highly productive site (Corradi *et al*. 2005) close to Cherskii in Sakha Republic. During three growing seasons, methane emission varied between 20 and 24 g(C) m^{-2}. After the experimental water table went down in the fourth year, methane emissions were close to zero.

Western Siberian lowlands constitute the most extensive wetland area in the world. Mires deeper than 0.5 m cover an area of 690 000 km^2 (Romanova *et al*. 1977). Several Russian studies were conducted in the western Siberian lowlands. Methane emissions were measured either by static chambers or by solving it from

concentration profile in the peat (e.g. Glagolev and Shnyrev 2007, Glagolev *et al*. 2008). Methane emissions from natural mires in the middle and southern taiga zones of Tomsk oblast were studied in the summer and autumn of 2006 (Glagolev and Shnyrev 2008). Methane fluxes (mg(C) m^{-2} h^{-1}) from different types of mires and different microelements of the surface topography were characterized by the following median values: 0.67 for a forested bog; 4.06 and 2.67 for hollows and elevations (hummocks) of an open (nonforested) mire, respectively; and 0.74 and 2.13 for hollows and ridges of a ridged bog, respectively.

Methane emission measurements in Khanty-Mansiysk district were conducted by Glagolev and Suvorov (2007). Naumov *et al*. (2007) measured summer average emissions of 5.6, 3.8 and 0.5 mg(C) m^{-2} h^{-1} from hollow, fen and ryam, respectively. Glagolev *et al*. (2008) made reference to measurements made in Noyabrsk in summer 1999, where the mean CH_4 emission from peat was 0.57 mg(C) m^{-2} h^{-1}. In the tundra subzone with a discontinuous permafrost layer near Pangody, the summer average CH_4 emissions were 0.25 and 1.2 mg(C) m^{-2} h^{-1}, in frozen palsa and oligotrophic hollow, respectively (Naumov *et al*. 2007).

CO_2 and CH_4 fluxes were measured from three small wetland lakes located in the middle taiga and forest tundra zones on West Siberian Lowlands (Repo *et al*. 2007). CH_4 ebullition was detected in two of the lakes. Total carbon evasion from the studied lakes during the active season was 23–66 g(C) m^{-2}, of which more than 90% was released as CO_2(C).

Glagolev *et al*. (2010b) calculated CH_4 emissions from mires of western Siberia by first compiling CH_4 emission factors for the various microsites in mires classified according to their nutrient status for different ecoregions and then combining the emission factors with land use information. This analysis gave total annual CH_4 emission from western Siberia to be 5.2 Mt(C).

Different ecosystems have specific net balances of greenhouse gases. Wetlands are characterized by methane emissions and, in stable conditions, net CO_2 uptake. Uplands are long-term net CO_2 sinks, but may loose large amounts of carbon during a disturbance event such as a forest fire. Climate change and direct human interventions perturb the long-term balance, which have prevailed during the past millennia. The environmental changes will be different in upland and wetland ecosystems and in the continuous and non-permafrost regions. The most visible changes will possible take place in the border zone where permafrost landscape elements disappear totally. The net GHG measurements by the micrometeorological method should span the dominant ecosystems and ecoregions in Siberia. More flux sites are needed especially in the intermittent permafrost zone, where hydrological and biological changes will be remarkable.

Atmospheric concentration measurements

The Voeikov Main Geophysical Observatory has monitored GHG concentrations in the Arctic since 1980s as part of the Global Atmosphere Watch programme of the World Meteorological Organization (WMO). Flask samples have been collected from Teriberka (69°12´N, 35°06´E) in the Kola Peninsula on the shore of Barents Sea and CO_2 has been analyzed since 1988 and CH_4 since 1999 (Paramonova *et al*. 2001). The data were submitted to the WMO World Data Centre for Greenhouse Gases (http://gaw.kishou.go.jp/wdcgg/). They have also reported to the WMO data centre a multiyear 1986–1993 dataset of CO_2 concentrations from Kotelny island (76°00´N, 137°50´E) in the Laptev Sea. Since 2003, CO_2 and CH_4 have been regularly analyzed from samples collected in Novyi Port in the north of the western Siberian lowlands. Zinchenko *et al*. (2008) and Reshetnikov *et al*. (2009) used these data together with additional measurements to estimate CH_4 emissions from natural gas production. Previously, Jagovkina *et al*. (2001) estimated that anthropogenic emissions in the summer are only about 10% of total wetland emissions in the area.

A measurement network was established in western Siberia within a Japan–Russia Siberian Tall Tower Inland Observation Network (Arshinov *et al*. 2009). The network consists of nine towers in steppe, taiga and extensive wetland environments. The top measurement heights vary between 43 and 85 m. CH_4 is measured by

a semiconductor sensor and CO_2 is measured by a non-dispersive infrared analyzer. According to Sasakawa *et al*. (2010), the main source of CH_4 concentration variations are wetlands in summer and fossil fuel emissions in winter.

Kozlova *et al*. (2008) described a tall (300 m) tower for CO_2, CO, CH_4 and O_2 concentration measurements at Zotino, in the boreal forest of central Siberia. Kozlova *et al*. (2008) quantified the oceanic component of the O_2 seasonal amplitude in this highly continental site. They also observed relatively high surface CO_2 emissions even during very cold season. More recently, a new low-maintenance system was set up (Winderlich *et al*. 2010). The data showed strong diurnal variations of CO_2 and CH_4 concentrations in the deep continental surface layer.

A series of continental scale snapshots of greenhouse, trace gas and aerosol concentrations were obtained using a laboratory wagon traveling along the Trans-Siberian railroad within the framework of the Trans-Siberian Observations Into the Chemistry of the Atmosphere (TROICA) project (Crutzen *et al*. 1998, Belikov *et al*. 2006, Kuokka *et al*. 2007, Tarasova *et al*. 2006, Vartiainen *et al*. 2007, Berezina and Elansky 2009, Elansky *et al*. 2007, Tarasova *et al*. 2009, Turnbull *et al*. 2009). During the midsummer, TROICA5 expedition found highest CH_4 emissions from wetlands in the western Siberian region (Oberlander *et al*. 2002). Outside urban areas, sources of elevated CH_4 concentrations were mire, forest fires and coal mining. Correlations between ^{222}Rn and CO_2 concentrations showed that highest soil respiration rates were in the Far East, where climate was warm and dry (Oberlander *et al*. 2002).

A network of atmospheric concentration observations around Siberia is needed for inversion estimates of GHG balances of Siberia. At least two sites should be located near the coast of the Arctic Sea, one in the eastern and one in the western part of the Siberia. Similarly, there should be monitoring stations in the southern border of the Taiga region. A station in the Ural mountains would help to separate Siberian sources and sinks from the forested areas of the European Russia. The Zotino tower in the continental central part of Siberia is most valuable for regional separation and for understanding vertical diffusion processes. More continuous atmospheric sites are needed to resolve sources and sinks in the non-permafrost, intermittent and continuous permafrost regions where environmental changes will be most likely different.

Aircraft observations

Airborne observations may cover extensive areas and, as such, are appropriate for Siberia. The observed atmospheric concentration is a sum of emissions from both natural and anthropogenic sources. If those need to be separated, one has to use other information such as tracers of known origin or isotopes (Lloyd *et al*. 2002, Ramonet *et al*. 2002, Paris *et al*. 2010a).

In the boundary layer above western Siberia, Tohjima *et al*. (1996) observed elevated methane concentrations originating from gas and oil production facilities and wetlands. Extensive air sampling over European Russia and Siberia was conducted within the Japan–Russia joint program, the Siberian Terrestrial Ecosystem–Atmosphere–Cryosphere Experiment (STEACE) in the summers of 1992, 1993, and 1994 (Nakazawa *et al*. 1997). These first airborne observations in the free troposphere and boundary layer showed large scale summertime CO_2 uptake by taiga forests, which was confirmed by inverse relationship between CO_2 and stable isotopes $\delta^{13}C$. High concentrations of CH_4 were observed over wetlands and big industrial areas, such as Moscow region. Small horizontal and vertical gradients of nitrous oxide suggest low emission rates.

The European project EuroSiberian CarbonFlux provided multi-year aircraft observations, which gave further insight into the seasonal cycles of GHG fluxes over European Russia and the central parts of Siberia (Levin *et al*. 2002). Lower troposphere GHG profiles over forested areas WNW from Moscow (Ramonet *et al*. 2002), the northern Urals (Sidorov *et al*. 2002) and central Siberia (Lloyd *et al*. 2002) revealed a significant increase in the seasonal amplitudes of CO_2 and $\delta^{13}C$-CO_2 in the free troposphere from western Europe to Siberia, where CO_2 seasonal amplitude was 15 ppm. The amplitude in the planetary boundary layer was even larger (25 ppm) and preceded about one month the free

troposphere cycle, demonstrating the strong CO_2 source-sink forcing by forests and wetlands over Russia. Styles *et al*. (2002) measured summertime CO_2 uptake and stable isotope discrimination on landscape level by the convective boundary layer (CBL) budget technique in a vegetated region in central Siberia. The source area analysis and covariance of GHG concentrations and isotopic signatures show that anthropogenic emissions are the major source of GHG concentration variations during winter (Levin *et al*. 2002). In the European Russia, anthropogenic emissions are the dominant source of concentration variations throughout the year (Ramonet *et al*. 2002).

Atmospheric inverse modeling is a tool for estimating the large-scale spatial GHG source and sink intensities. These calculations use *a priori* emission intensity maps, an atmospheric transport model, and GHG observations from a network, which are mostly surface observations. A major source of uncertainty in these calculations is a poor simulation of transport and mixing processes over the continents (Gurney *et al*. 2002). The vertical diffusion of depleted surface CO_2 concentrations in summer under convective conditions and of elevated concentrations in winter below strong inversion are not simulated accurately, which results in erroneous horizontal advection fields. The problem is stressed by the fact that inversions rely heavily on surface observations and, thus, on the simulation of exchange processes between the surface and the free troposphere.

For advancing our understanding of diffusion and advection processes over Siberia, a joint French–Russian research program Airborne Extensive Regional Observations in Siberia (YAK-AEROSIB) was organized, including aircraft measurement campaigns (Paris *et al*. 2010b). Observations of CO_2 and other trace gases on Trans-Siberian flights revealed long-range transport of anthropogenic pollutants by synoptic systems (Paris *et al*. 2008). One of the pathways of anthropogenic emissions from the industrialized Europe was to the Siberian lower troposphere. Emissions from China were more often advected to the upper troposphere over Siberia. A notable source of elevated concentrations of CO_2 and other trace gases was the biomass burning in Kazakhstan. In summer, depleted CO_2 concentrations were observed when air masses had stayed in the boundary layer over the boreal and Arctic Siberia (Paris *et al*. 2010a). In summary, airborne observations show the complicated atmospheric advection of elevated concentrations from various anthropogenic emission categories and biogenic activity during the growing season. Correct simulation of these processes is a challenge. The fact that in the continental part of Siberia atmospheric transport models have difficulties in properly calculating mass transfer from the surface to the free troposphere under very stable boundary layer conditions in winter and convective conditions in summer (Stephens *et al*. 2007) strongly supports *in-situ* monitoring of lower tropospheric concentrations by aircraft.

Atmospheric column observations of CO_2 and CH_4

Total atmospheric column concentrations are measured from high-resolution solar spectra. These data are useful for long-term trends and for atmospheric models because they provide information on the concentrations above the boundary layer (Kashin *et al*. 2004). There are currently regular spectroscopic column methane observations at three sites in Russia: at the Research Institute of Physics of the St. Petersburg State University, at the IAP Zvenigorod Scientific Station in the vicinity of Moscow and at the Institute of Experimental Meteorology in Obninsk, 100km SW from Moscow (Makarova *et al*. 2009). Kashin *et al*. (2008) reported on the long-term (beginning in 1980) CO_2 column observations in Kyrgyzstan. Recently, very accurate instrumentation has been obtained for these measurements in St. Petersburg (Poberovskii *et al*. 2010).

Total Carbon Column Observing Network (TCCON) of ground-based Fourier Transform Spectrometers, which record direct solar spectra in the near-infrared, uses standard setup for precise column-averaged abundances of atmospheric constituents, including CO_2, CH_4, N_2O, HF, CO, H_2O, and deuterated water (HDO) (Geibel *et al*. 2010, Wunch *et al*. 2011, http://www.tccon.caltech.edu). The network provides

an essential validation resource for the Orbiting Carbon Observatory and GOSAT (Greenhouse gases Observing SATellites) satellites which measure total concentrations from space. One of the ground-based stations is starting operation in Yekaterinburg.

Remote sensing observations of column total concentrations of CO_2 and CH_4 are becoming available from a Japanese GOSAT (Yokota *et al*. 2009) and a future OCO-2 satellites (Crisp *et al*. 2004) which use solar-reflected near-infrared radiation for retrieving the concentrations. The aim of these missions is to use global column concentration observations together with atmospheric transport fields and inversion models to obtain regional source and sink maps. Indeed, Chevallier *et al*. (2009) estimated that GOSAT observations should significantly improve our knowledge of the CO_2 surface fluxes over terrestrial vegetated region, even at a scale of a week and of a few hundred kilometers. The positive impact depends on the accuracy of the satellite product and the quality of the atmospheric transport modeling (Chevallier *et al*. 2010). The accuracy of the GOSAT products is still developing. In high northern latitudes, high solar zenith angle restricts the seasonal period of retrievals and the accuracy of the retrievals. A necessary condition for high quality retrievals is a cloudless sunny weather that significantly reduces the number of observations in boreal and arctic regions. For the development of the accuracy and validation of the space observation products, ground-based TCCON network should be extended to span geographical area and environmental conditions of Siberia. TCCON observations are more accurate than will be the space observations. Column measurements in inflow and outflow areas of Siberia could help in improving regional source and sink estimates in spite of the fact that it is difficult to resolve a very small latitudinal total concentration gradient embedded in a strong meridional gradient (Keppel-Aleks *et al*. 2011).

Future issues and Pan Siberian Experiment (PSE)

Although the overview presented in the previous section shows increasing efforts to obtain proper data and information from sources, sinks and concentrations of greenhouse gases and aerosols over Siberia, continuous, comprehensive and integrative measurements are still lacking; e.g. simultaneous continuous measurements of CO_2, methane, ozone, VOCs and aerosols, together with biosphere–atmosphere exchange, are missing. Nowadays, information is mainly coming via measurement campaigns, not via continuous monitoring. Actually, continuous and comprehensive investigations need be performed at different ecosystems. In practice this means (a) updating existing research stations and establishing new ones, (b) utilizing the data in different models via data handling and processing; (c) improving data quality; (d) ensuring the access to the data; (e) developing new instrumentation when needed; (f) utilizing satellite data for wider geographical areas all around Siberia; and (g) utilizing observations from all scales to develop and evaluate process-models of biota-atmosphere interactions.

Due to the significance of boreal forest and arctic area from the climate change point of view, and due to the lack of detailed information, more investigations are needed. Based on the overview presented in the previous section, several scientific questions related to Siberian aerosols and greenhouse gases and their connection with future climate change can be identified:

- How fast will the permafrost thaw proceed?
- How will the thawing permafrost affect hydrology, carbon fluxes and methane emissions?
- How rapidly will the permafrost melt? How will the melting permafrost affect methane emissions and net CO_2 uptake?
- How will the cryosphere change over the next decades?
- What are the foreseen land use changes in Siberia and other Euro-Asian boreal forest areas, and how would that affect exchanges of greenhouse gases and aerosol precursors?
- What are the present BVOC emissions and how will they change during the next 10 and 30 years over Siberia and other Euro-Asian boreal areas?
- What are the connections between ecosystem carbon cycle, the chemistry of biogenic

volatile organic compounds, ozone chemistry and the formation and growth of aerosol particles?
- What are the regional and global climate and air quality effects of biomass burning in Siberia? How do burning regimes change in response to land use change and to climate change?
- How could areas over Siberia especially sensitive to climate change be identified?
- How do the above processes and problems relate to spatial and temporal distribution of basic features of atmospheric planetary boundary layer (first of all, its height) over Siberia and other northern Euro-Asian areas?

Addressing these scientific questions and the related societal effects in changing climate requires inter-, multi- and cross-disciplinary research, along with a high level of technological and scientific innovation and expertise in the areas of chemistry, physics, biology and meteorology. As a solution, we propose a so-called Pan Siberian Experiment (PSE), which would investigate the concentrations and fluxes of aerosol particles, trace gases and greenhouse gases, including their interlinks and related processes. An important part of the PSE would be a network of research stations from Scandinavia to China with a continuous, comprehensive science program. Continuous data would also be obtained from satellites, and it would be complemented with *in-situ* measurement data from aircraft, trans-Siberian railway and research vessels. Several targeted field experiments would have to be performed as well to investigate processes in more detail. The station network would include several SMEAR-type stations (*see* Hari and Kulmala 2005, Hari *et al*. 2009) and GAW stations. It would be crucial to have one supersite in all major ecosystem areas (Fig. 1), which in practice would mean a station in every 2000–3000 km.

Our scientific plan in PSE is designed as a research chain that aims to advance our understanding of climate and air quality through a series of connected activities beginning at the molecular scale and extending to the regional and global scale (*see* Kulmala *et al*. 2009). Fundamental aerosol and carbon cycle processes need to be understood in order to quantify radiative properties and the influence of aerosols on cloud microphysics and dynamics at the scale of individual clouds, and to understand changes in carbon uptake dynamics. At larger scales, advances in our understanding of boundary layer meteorology are needed to understand atmospheric aerosol transport, trace gas (e.g. CO_2, methane, N_2O, O_3, SO_2, NO_x, VOCs) and water vapor exchange and deposition processes. Boundary layer studies form a link to regional-scale processes and further to global-scale phenomena. In order to be able to simulate global climate and air quality, the most recent progress in this chain of processes must be compiled, integrated and implemented in Climate Change (CC) and Air Quality (AQ) numerical models via novel parameterizations. Anthropogenic effects on ecosystems and cryosphere will be investigates, including land use changes.

The *in-situ* field experiments would be carried out on ground-based stations, aircraft and ships, in addition to using existing data sets and archives. They would provide data on (i) short-lived pollutant concentrations and greenhouse gas concentrations in the air, biosphere and on snow and ice surfaces, (ii) seasonal evolution of terrestrial and oceanic snow and ice cover, (iii) surface radiation and heat exchange, (iv) fluxes of key species, (v) cloud properties, and (vi) relevant meteorological variables. The observation network would consist of existing intensive stations and their measurement programs, including quality analysis and data dissemination procedures developed in other projects. These would be updated with additional instruments addressing the research questions. To ensure long-term sustainability and comparability, these measurements would be connected to international networks wherever possible.

The basic observations would be performed within the atmospheric planetary boundary layer (PBL), that is, the strongly turbulent layer immediately affected by dynamic, thermal and other interactions with the Earth's surface. It essentially differs in nature from the free atmosphere. The latter is only weakly turbulent (because of its very stable stratification) and experiences the abovementioned impacts indirectly, in an aggregated form thorough the PBL integration-and-coupling mechanisms. To some extent, the PBL

upper boundary plays the role of a lid preventing dust, aerosols, gases and other admixtures released from ground sources to efficiently penetrate upwards, thus blocking them within the PBL (Zilitinkevich 1991, Zilitinkevich *et al*. 2007). Furthermore, perturbations of the heat budget at the Earth's surface are almost completely absorbed by the PBL through the very efficient turbulent heat-transfer mechanism. By these means, the PBL height, varying from a few dozen meters to a few kilometers, to a large extent controls dispersion and transport of atmospheric admixtures, extreme cold and heat, local amplification of global warming, and consequences of the land-use changes. Apparently, it affects terrestrial ecosystems, permafrost and cryosphere as a whole, not to mention the urban environment. The most sensitive are shallow stable PBLs typical of Siberian winters and the nighttime in all seasons (Zilitinkevich and Esau 2009). Comprehensive inventory of the PBL height over Siberia would be addressed in a sub-project within PSE, involving numerous Russian institutions.

Remote sensing with satellites would be used to retrieve information on e.g. aerosol and cloud properties, UV radiation, trace gas concentrations, biosphere, snow cover, ice extent, surface albedo, and top of atmosphere radiation. Many of the relevant quantities are measurable only from satellites. Satellites would complement *in-situ* and modeling data related to chemistry, aerosols, clouds and their interactions, especially for the spatial and temporal distribution pattern of relevant quantities. Derived data would be used to evaluate large-scale models simulating important Earth System components.

Past variations in climate and corresponding forcing agents would be revealed by analysis of firn and ice cores in glaciers and ice sheets. Interpreting ice cores correctly is probably the area where atmosphere and cryosphere scientists have the greatest potential of working together, since both fields are needed for interpreting the ice core data and for utilizing them in Earth System Modeling.

Finally, a variety of modelling frameworks would be used to identify the importance of different processes on short-to-moderate temporal and spatial scales, to extrapolate measurement results and to predict future scenarios. The models, like observations, would cover different spatial and temporal scales, and they would be implemented in a hierarchical structure. The applied models would range from detailed process models and radiative transfer models up to regional and global chemical transport and climate models and Earth System models.

The proposed PSE plan is evidently highly ambitious. However, with well-coordinated cooperation using the already existing tools in an inventive way, it could be considered achievable (*see* also Groisman and Soja 2009, Gordov and Vaganov 2010). The results obtained from such an experiment would be highly useful for a broad scientific community.

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