Overview of the atmospheric research activities and results at Pallas GAW station

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Finnish Meteorological Institute's air quality measuring station at Pallas, in northern Finland, has been part of the Global Atmosphere Watch programme of the World Meteorological Organization since 1994. Tropospheric air composition and meteorological measurements are made at four different locations. Measured components include greenhouse and reactive gases, volatile organic compounds, chemistry of precipitation and aerosols, radon, carbon dioxide flux, meteorological parameters and a number of different aerosol characteristics. Pallas has proven to be a good site for measuring continental background air. This paper gives a general overview of the Pallas GAW station measurement sites and methods, and extensive overview of the scientific results.

Introduction

Human activities have an increasing influence on the global atmosphere, including the depletion of the stratospheric ozone layer, increases in tropospheric ozone, acid deposition, rising greenhouse gas concentrations, and changes in the radiative balance of the earth-atmosphere energy system. To understand the mechanisms behind these changes, and to be able to predict future atmospheric states, reliable and long-term data and information on chemical composition and related physical characteristics of the atmosphere is required on global and regional scales. World Meteorological Organization's (WMO) Global Atmosphere Watch (GAW) programme was established in 1989 and its objective is to provide these data. Monitoring priorities have been given to greenhouse gases, ozone and ultraviolet radiation, certain reactive gases and the chemistry of precipitation (World Meteorological Organization 2001).

The GAW monitoring network consists of 22 global stations and ca. 300 regional stations. Global stations are situated in remote locations, have very low (background) levels of pollutants that are representative of large geographical areas, and continuously measure a broad range of atmospheric parameters. Finnish Meteorological Institute's (FMI) measuring stations at Pallas

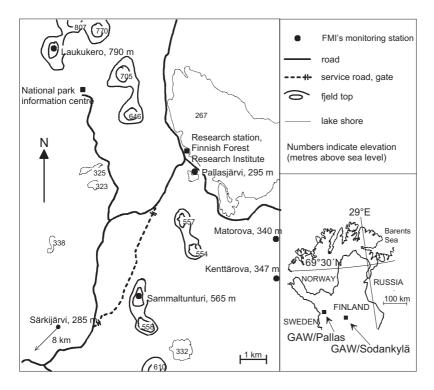


Fig. 1. Location of Pallas and Sodankylä GAW stations (lower right), and location of measuring stations at Pallas (left).

and Sodankylä in northern Finland together form one of these global stations. Pallas-Sodankylä was incorporated into the GAW programme in 1994.

Upper-air soundings, including ozone, spectral UV measurements, climatological and other meteorological measurements, are made at the FMI's Arctic Research Centre in Sodankylä, while tropospheric air composition and related meteorological measurements are made at Pallas. The distance between the two sites is 125 km (Fig. 1). This paper gives a detailed description of the Pallas site, its measuring programme and instrumentation, together with an overview of the main results obtained.

Methods and materials

Experimental sites and measuring programme

Pallas is in the subarctic region at the northernmost limit of the northern boreal forest zone. Surrounding forest is mixed pine, spruce and birch. The area has four separate stations within 12 km of each other. Two of the stations are located inside the Pallas-Ounastunturi National Park, and the other two in Finnish Forest Research Institute's research forests. The distance to the nearest town, Muonio, with some 2500 inhabitants, is 19 km to the west. The area has no significant local or regional pollution sources.

The stations at Pallas have different measuring programmes. Sammaltunturi is the main station with the largest programme. Matorova station was built mainly due to the conditions being too harsh at Sammaltunturi during wintertime for filter sample collection. A 20 m high tower was built at Kenttärova for micrometeorological flux measurements. Laukukero is a weather station on top of a high fjeld (arctic round topped hill), and its results are used together with weather data from the other three stations to obtain information about the structure of the boundary layer in the area. A forest weather station, Pallasjärvi (68°01'05''N, 24°10'04''E, 303 m above sea level) was in operation between 1996-2002 in the same area. In addition, a station collecting deposition samples, Särkijärvi (67°55'N, 23°55'E, 260 m above sea level), 10 km WSW of Sammaltunturi, was in operation from 1991 to 1999. All the coordinates are given in WGS 84 geodetic system.

Sammaltunturi (67°58′24′′N, 24°06′58′′E)

The Sammaltunturi station resides on top of the second southernmost fjeld at an elevation of 565 m a.s.l. in a 50 km long north–south chain of fjelds. The tree line is some 100 m below the station. The vegetation on the fjeld top is sparse, consisting mainly of low vascular plants, moss and lichen. The highest fjelds in the chain are from 600 to 800 m a.s.l. Otherwise the region is hilly (250–400 m a.s.l.), forested and partly swampy with some rather large lakes (ca. 250 m a.s.l.). In N, NE and SSW there are fjelds within 3–6 kilometres from the station with heights from 600 to 800 m a.s.l. The sectors 180–330° and 100–130° are very open. The station's WMO index number is 05821.

A new 120 m² station building was taken into use in July 2001. Before that the instruments were in an old building, where monitoring activities began in 1991, 40 m west of the new station. The new station has a common sampling line, from which nearly all instruments take their sample air. The sampling line is made from 56 mm internal diameter acid resistant stainless steel tubing, through which a continuous flow of 90 m³ h⁻¹ (corresponding to flow speed 10 m s⁻¹) is maintained. The residence time within the sampling line is less than two seconds, and the exhaust and building ventilation air is led ca. 50 m away from the station. The warmed inlet is seven meters above the ground. The station has also a common vacuum line, so that analysers do not have to have separate vacuum pumps. The old station had a similar sampling and vacuum line arrangement. The station's measuring programme is presented in Table 1.

Matorova (68°00'00''N, 24°14'24''E)

Matorova lies six kilometres ENE of Sammaltunturi at an elevation of 340 m a.s.l. It is situated on top of a small hill covered by coniferous forest in

Component	Measurement method	Frequency	Period	
Ozone	UV absorption	Continuous	1995–	
Sulphur dioxide	UV fluorescence	Continuous	1995–	
Nitrogen oxide + dioxide	Chemiluminescence	Continuous	1995, 1999–	
Carbon dioxide	NDIR analyser	Continuous	1996-	
Carbon monoxide	Reduction gas analyser	Continuous	2002-	
Aerosol number concentration	Da >10 nm, Condensation Particle Counter	Continuous	1996-	
Aerosol number concentration	Da > 0.5 μm, Laser Particle Counter	Continuous	1996-	
Radon-222 (progeny)	Filter collection + beta counting	Continuous	1995–	
Radon-222	Delay chamber – alpha counting (EML)	Continuous	2002-	
Black carbon	Aethalometer, light absorption	Continuous	1996-	
Aerosol scattering coefficient	Three wavelength integrating nephelometer	Continuous	2000-	
Aerosol size distribution	Differential mobility particle sizer	Continuous	2000-	
Volatile organic compounds	Flask sampling, GC analysis	2/week	1994—	
CH₄, N₂O, SF ₆	NOAA flasks, GC analysis	1/week	2002-	
Carbon monoxide, hydrogen	NOAA flasks, reduction gas analyser	1/week	2002-	
Carbon dioxide	NOAA flasks, NDIR analyser	1/week	2002-	
Wind speed and direction	6 m above ground, cup anemometer + vane	Continuous	1995–	
Temperature + rel. humidity	4 m above ground, Pt100 + HUMICAP	Continuous	1995–	
Pressure	2 m above ground, Vaisala DPA21	Continuous	1995–	
Visibility, present weather	Vaisala FD12P present weather sensor	Continuous	1995–	
Global radiation	Pyranometer	Continuous	1995–	
PAR	Photovoltaic detector	Continuous	1995–	

Table 1. Summary of measurements at Sammaltunturi station. Abbreviations: EML = Environmental Measurement Laboratory, USA; NOAA = National Oceanic and Atmospheric Administration, USA.

the middle of a ca. 100×100 -m clearing. The station is used mainly to collect deposition, gas and aerosol samples (Table 2). Special attention was paid to the selection of building materials for the station, so that it would be suitable for collecting samples for trace metal, mercury and persistent organic pollutant analysis. For the same reason motor vehicle use is limited to the minimum. Also walking trails are made of wood, so that the surface soil is not exposed to wind-driven erosion, which could possibly cause resuspended material to contaminate the samples. There is a level II intensive monitoring site of the International Co-operative Programme on the Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) very near the Matorova station. The station also participates in Arctic Monitoring and Assessment Programme (AMAP).

Kenttärova (67°59′14′′N, 24°14′36′′E)

The station lies at an elevation of 347 m a.s.l on a hill top plateau, 1.4 km south of the Matorova station. The hill is ca. 60 m above the surrounding plains. The 20-m high measurement tower is situated in a Norway

spruce forest (Hylocomium-Myrtillus type). The mean tree density of the stand is 1050 stems per hectare and the dominant tree height is 13 m. The age of the trees varies from 70 to 160 years. The stand is naturally regenerated, but some forest management has taken place, as most of the downey birch (Betula pubescens) was cut down in 1968-1969. The total leaf area index of 6 m² m⁻² was estimated for the site by using the allometric functions of Marklund (1987, 1988) and the specific leaf dry-weight information of Steinbrecher et al. (1999) determined at the Pallas area. The station's WMO index number is 05824. Table 3 presents the measuring programme at the station.

Laukukero (68°03'48''N, 24°02'02''E)

Laukukero automatic weather station is on top of a fjeld (765 m a.s.l.), 10.6 km NNW of the Sammaltunturi station. The station has standard weather measurements: wind, temperature, relative humidity and atmospheric pressure (Table 4). The WMO index number of the station is 05820.

 Table 2. Summary of measurements at Matorova station. Abbreviations: IVL = Swedish Environmental Research

 Institute, FEA = Finnish Environment Agency.

Component	Measurement method	Frequency	Period	
Sulphate	Filter	Daily	1996–	
Sulphur dioxide	Impregnated filter	Daily	1996—	
Nitrate + nitric acid	Filter + impregnated filter	Daily	1996—	
Ammonium + ammonia	Impregnated filter	Daily	1996—	
Trace metals	Filter	Weekly	1996–	
Mercury, particulate	Filter (IVL)	Weekly	1996—	
Mercury, gaseous	Goldtrap (IVL)	2 days/week	1996—	
Persistent organic pollutants	Filter + polyurethane foam (IVL)	1 week/month	1996—	
pH, conductivity, major ions	Precipitation, bulk collector	Monthly	2000-	
Trace metals	Precipitation, bulk collector	Monthly	1996—	
Mercury	Precipitation, bulk collector (IVL)	Monthly	1996—	
Persistent organic pollutants	Precipitation, bulk collector + polyurethane foam (IVL)	1 week/month	1996—	
Persistent organic pollutants	Precipitation, bulk collector (FEA), summer	Monthly	1995–	
Radon-222 (progeny)	Filter collection + beta counting	Continuous	1998-2001	
Aerosol size distribution	Differential mobility particle sizer	Continuous	1999–2001	
Wind speed and direction	14 m above ground, cup anemometer + vane	Continuous	2000-	
Temperature + rel. humidity	2 m above ground, Pt100 + HUMICAP	Continuous	2000-	
Pressure	2 m above ground, Vaisala DPA21	Continuous	2000-	
Precipitation amount	1 m above ground, rain gauge	Weekly	1998–	

Experimental setup

Weather

All the stations are equipped with Vaisala Milos 500 automatic weather stations. The wind speed is measured with a heated cup anemometer and direction with a heated wind vane. Temperature is measured with Pt100 sensors and relative humidity with a Vaisala sensor (HUMICAP). Barometric pressure is measured with Vaisala BAROCAP sensors. In addition, Sammaltunturi station has a Vaisala FD12P present weather sensor, which measures visibility and precipitation intensity and type. Data is saved as one minute means.

Carbon dioxide

Carbon dioxide (CO_2) measurements started at Sammaltunturi station in October 1996 with a system built and installed by Air Quality Research Branch/Meteorological Service of Canada (AQRB). It was based on a Unor 6N non dispersive infrared (NDIR) analyser, and used one reference gas, two station standards and a target gas. The system operated in five minute cycles, and only the last one minute mean value of every cycle was saved. Station standards were measured once an hour, and reference and target gases every five hours. All the gases were supplied and calibrated by AQRB. Air was drawn through a separate sampling tower from 7 m above the ground with a diaphragm pump. The sample air was dried with a chemical dryer (magnesiumperchlorate). The flow rate was 100 ml min⁻¹ for the sample and 10 ml min⁻¹ for the reference gas. The system was operational until March 1998.

FMI installed its own CO_2 measurement system in July 1998. The original system was based on a LI-6262 (Li-Cor, Inc.) CO_2/H_2O NDIR analyser, which was replaced with a LI-6252 CO_2 NDIR analyser in 2000. The system uses three station standards and a reference gas. A sample is taken from a common sampling line with a diaphragm pump and predried with

Table 3. Summary of measurements at Kenttärova station (established in November 2002).

Component	Measurement method	Frequency	Period	
Carbon dioxide and H ₂ O flux	23 m above ground, NDIR analyser	Continuous	2002–	
3-dimensional wind	23 m above ground, sonic anemometer	Continuous	2002-	
Wind speed and direction	22 m above ground, cup anemometer + vane	Continuous	2002-	
Temperature + rel. humidity	3 m above ground, Pt100 + HUMICAP	Continuous	2002-	
Temperature + rel. humidity	18 m above ground, Pt100 + HUMICAP	Continuous	2002-	
Temperature + rel. humidity	21 m above ground, Pt100 + HUMICAP	Continuous	2002-	
PAR, SW and IR radiation 22 m above ground, radiometer		Continuous	2002-	
Reflected PAR, SW and IR 22 m above ground, radiometer		Continuous	2002-	
Soil temperature	–0.05 m, Pt100	Continuous	2002-	
Soil temperature	-0.20 m, Pt100	Continuous	2002-	
Soil temperature	–0.50 m, Pt100	Continuous	2002-	
Soil humidity -0.10 m, Delta-T Theta probe		Continuous	2002-	
Soil humidity	–0.40 m, Delta-T Theta probe	Continuous	2002-	
Soil heat flux	–0.10 m, heat flux plate	Continuous	2002–	

Table 4. Summary of measurements at Laukukero station.

Component	Measurement method	Frequency	Period
Aerosol size distribution	Differential mobility particle sizer	Continuous	2002–
Wind speed and direction	11 m above ground, cup anemometer + vane	Continuous	1996–
Temperature + rel. humidity	3 m above ground, Pt100 + HUMICAP	Continuous	1996–
Pressure	2 m above ground, Vaisala DPA21	Continuous	1996–

an electric gas cooler to -30 °C, after which it is further dried chemically with magnesiumperchlorate. The flow rates through the analyser are 100 ml min⁻¹ and 10 ml min⁻¹ for the sample and reference gases, respectively. Flows are regulated with mass flow controllers. Station standards are measured every 2.5 hours, and in addition the reference gas is measured every 7.5 hours. In August 2000 FMI obtained its own set of seven cylinders of WMO/Central CO, Laboratory (CCL) standard gases. The station standards and the reference gas are calibrated against the CCL standards every two to three months at the station with the same measuring system. Results are saved as one minute mean values with minimum, maximum, deviation and median values.

At the Kenttärova station CO₂ and sensible and latent heat fluxes are measured using eddy covariance (EC) technique. The instrumentation includes a SWS-211 (Applied Technologies, Inc.) three-axis sonic anemometer and a LI-7000 (Li-Cor, Inc.) closed path CO₂/H₂O analyser. The sonic anemometer is mounted at a height of 23 m close to the mouth of the inlet tube of the LI-7000. The heated inlet tube is 8 m in length and has an inner diameter of 3.1 mm. Turbulent flow is achieved in the tube by using a flow rate of ca. 6 l min⁻¹. A sampling rate of 10 Hz is used for the EC system. The measuring system has been described in more detail by Aurela et al. (2001). An extensive set of additional meteorological measurements are conducted at the site. Temperature and humidity profiles in air and in soil and the photosynthetic photon flux density (PPFD) are essential variables in explaining the physiological functioning of the forest ecosystem. The latent and sensible heat fluxes measured by EC are complemented by the measurements of net radiation and soil heat flux in order to study the energy budget.

Ozone, sulphur dioxide, nitrogen oxide and nitrogen dioxide

Concentrations of NO and NO₂ play a key role in the photochemical processes. Accurate measurement of their concentration in remote areas is, however, challenging. In 1998, NO and NO₂ were measured with an ECOPHYSICS CLD780TR NO monitor and a PLC762 photolytic NO₂ converter. PLC762 converts selectively NO₂ to NO. NO was calibrated automatically adding ppm level NO calibration gas (by Messer-Griesheim) to the sampling line to get a 4.8 ppb increase to the observed NO concentration. This gives the ratio between photomultiplier counts and NO concentration. In May–June 1998, NO₂ mode was not active but the system was continuously measuring NO. NO₂ concentrations were calculated using photostationary state assumption (e.g. Ridley *et al.* 1993) and measured JNO₂ photolysis frequencies and ozone concentrations.

Since September 1999 nitrogen oxide concentrations have been measured with a chemiluminescence analyser with molybdenum converter, Thermo Environmental Instruments (TEI) 42 CTL. In addition to NO₂, the molybdenum converter converts also other oxidised nitrogen species (most importantly HNO₂ and organic nitrates such as PAN) to NO (Williams et al. 1998, Thielmann et al. 2002). Thus the "NO₂" concentration measured by TEI42CTL is actually a sum of all oxidised nitrogen species, NO₂. Usually in remote areas NO₂ is only a small fraction of NO_v (Wang *et al.* 2003). Due to very low NO_x concentration levels at Pallas, especially during summer, the results are most of the time below the instrument's detection limit, which is according to manufacturer 50 ppt.

Ozone (O_3) is measured with instruments based on measuring UV absorbance at the wavelength of 254 nm. There are two separate ozone analysers running in parallel (Thermo Environmental Instruments 49C and Dasibi 1008). Only the data from the main analyser (TEI 49C) is reported, and the other instrument is used for backup/quality control purposes. Sulphur dioxide (SO₂) analysis is based on measuring UV fluorescence. The instrument used is Thermo Environmental Instruments 43S.

All these measurements are made at the Sammaltunturi station. Data is collected with a data acquisition system at one minute intervals. All the analysers are calibrated four times a year, ozone monitors against a portable calibrator, SO_2 and NO_2 (i.e. converter efficiency) against a permeation tube and NO with a calibration gas cylinder. The calibrators are in turn calibrated in FMI's FINAS-accredited calibration laboratory.

Aerosol measurements

Total aerosol number concentration is measured with a TSI Condensation Particle Counter (CPC) model 3010, which measures aerosol particles with aerodynamic diameter larger than ca. 10 nm. Number concentration is also measured with a Laser Particle Counter (LPC, TSI 7550), which counts aerosol particles with aerodynamic diameter larger than 0.5 μ m. The cut-off size for large particles due to inlet and sampling line is 4–10 μ m depending on wind speed.

Aerosol scattering and backscattering coefficients are measured with a three wavelength integrating nephelometer, TSI 3563. Flow rate through the instrument is ca. 25 m³ h⁻¹, and it is calibrated twice a year with pure CO_2 and clean air.

Particle size distribution is measured with a Differential Mobility Particle Sizer (DMPS). DMPS is built up with a 28-cm long Hauketype differential mobility analyser with a closed loop sheath flow arrangement and a CPC (TSI model 3010). The measured particle size range is from 7 to 500 nm, which is divided into 30 discrete bins. One particle size spectrum takes around five and a half minutes to measure (Komppula *et al.* 2003). A second DMPS system has been running at Matorova from April 2000 to February 2002, and at Laukukero from March 2002.

Aerosol black carbon is measured with an aethalometer (Magee Scientific). The instrument measures light absorption of aerosol collected onto a filter tape. The data collection period for all the above mentioned instruments is five minutes, and they are placed at the Sammaltunturi station.

Radon

The radioactive radon isotope ²²²Rn is measured at the Sammaltunturi station with two different instruments. These instruments have a common sampling line and inlet, with a flow rate of ca. $24 \text{ m}^3 \text{ h}^{-1}$. FMI's system is based on collecting aerosol continuously onto a filter and measuring the beta activity on it. The hourly mean values are calculated by assuming that the activity on the filter is from short-lived ²²²Rn progeny, and that no artificial activity is present (Paatero *et al.* 1998).

The other radon instrument is from the Environmental Measurements Laboratory (EML), U.S. Department of Energy. EML's instrument is based on the two-filter method, in which air is filtered (removing aerosol bound radioactivity) entering a decay volume (500 litres), and the short-lived radon progeny produced in the decay volume is collected onto a second filter. After a one-hour collection period this filter is moved to a counting location, where alpha particles resulting from further decay are counted with a scintillation detector.

Concentration of short-lived radon progeny nuclides in precipitation was measured with an automatic precipitation gamma analyser at Matorova in 1998. This was used to find out what meteorological parameters have influence on scavenging efficiency of the radon daughter nuclides (Paatero 2000).

Volatile organic compounds

Air samples are collected at Sammaltunturi station twice a week on Mondays and Fridays for volatile organic compound (VOC) analysis. The samples are collected to evacuated 0.8 litre stainless steel canisters with a portable pump, which is taken outside and upwind of the station. Two flasks are collected at a time.

Canister samples are analysed using a gas chromatograph (GC) equipped with a flame ionisation detector (FID) and Al₂O₂/KCl PLOT column (50 m, i.d. 0.32 mm). Prior to analysis samples are passed through a stainless steel tube (10 cm \times 1/4'') filled with K₂CO₃ and NaOH in order to dry them. Air samples are concentrated in two liquid nitrogen traps. The first trap is a stainless steel loop $(1/8'' \times 125 \text{ cm})$ filled with glass beads, while the other one is a capillary trap. Calibration is performed using a gas-phase standard from NPL (National Physical Laboratory, UK) including 30 hydrocarbons at concentration levels of 1-10 ppb. The detection limits for individual compounds are varying from 3 to 11 ppt.

Inorganic compounds and ions

Filter samples are collected at Matorova station on a daily basis. The filter pack method is used to collect gaseous sulphur dioxide, nitric acid and ammonia as well as particle bound sulphate, nitrate and ammonium. The filters are extracted with 10 ml of ultra pure water, and the extracts are analysed with ion chromatography (IC). Since the filter pack sampling method does not separate completely the gaseous nitric acid from the particle bound nitrate or the gaseous ammonia from the particle bound ammonium, the results are given as the sum of these components, $HNO_3(g) + NO_3^-(p)$ and $NH_3(g) + NH_4^+(p)$.

Weekly total (wet + dry) deposition samples are collected at Matorova station with a 0.04 m² bulk deposition collector. The samples are analysed at FMI's laboratory with ion chromatography for anions Cl⁻, NO₃⁻ and SO₄²⁻, and cations Mg²⁺, Ca²⁺, NH₄⁺, Na⁺ and K⁺. The electrical conductivity and pH of the precipitation samples are also measured, and H⁺ concentrations are calculated from pH values. From 1995 to 1999 the precipitation samples were collected at Särkijärvi (16 km WSW of Matorova).

Monthly precipitation and weekly aerosol samples are collected for trace element analysis using ICP-MS. These elements include Zn, Pb, Cu, Cd, Cr, Fe, Ni, Mn, V, Al, Co and As. Samples for persistent organic pollutants and mercury analysis are collected for the Swedish Environmental Research Institute, IVL (Bidleman *et al.* 2002, Wängberg *et al.* 2002).

Other measurements

Sammaltunturi station has participated in National Oceanic and Atmospheric Administration's (NOAA/CMDL/CCGG, U.S. Department of Commerce) Cooperative Air Sampling Network since the beginning of 2002. A pair of glass flasks is collected at the station once a week. The NOAA sampler has its own sampling line and heated inlet. The samples are analysed by NOAA in Boulder for CO₂, CH₄, CO, H₂, N₂O and SF₆, and by INSTAAR (Stable Isotope Laboratory at the University of Colorado) for stable isotopes of CO₂. Carbon monoxide measurements were started in 2002 at Sammaltunturi station based on a reduction gas analyser (Trace Analytical Kappa5). The instrument is running in five minute cycles, with a working standard measured every hour and three calibration gases once a day.

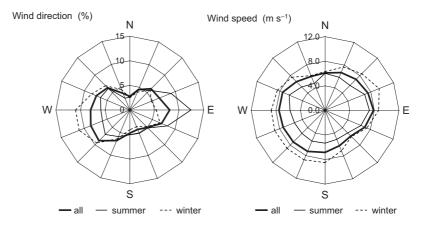
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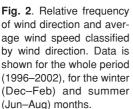
Climatology

Due to its northern location the Pallas area experiences ca. three and a half week long polar night in winter and seven week polar day in summer. The ground is covered with snow from the middle of October to late May (ca. 220 days), maximum snow cover depth (ca. 0.9 m) occurs during April. Annual amount of precipitation at the Pallas region is ca. 500 mm.

Average, minimum and maximum values for meteorological parameters are presented in Table 5 for the Sammaltunturi station for the years 1996-2002. Due to its elevation, Sammaltunturi station is from time to time within cloud cover. This happens most often during autumn, when nearly 40% of time the visibility is less than 1 km. The clearest season is summer, when the corresponding percentage is 10%. The wind speed is rarely (1.6% of the time) less than 1.0 m s⁻¹. However, there is a clear difference between winter (Dec-Feb) and summer (Jun-Aug): western winds are prevailing and wind speed is higher during winter (average wind speed 8.1 m s⁻¹, speed < 1.0 m s⁻¹ 0.7% of the time). In summer eastern winds are prevailing and the wind speed is lower (average 5.8 m s⁻¹, speed < 1.0 m s⁻¹ 2.2%), see Fig. 2. Bimonthly diurnal variation of several meteorological parameters are presented in Fig. 3. Atmospheric pressure and dewpoint temperature do not have a clear diurnal cycle, other parameters have, especially during summer.

The small seasonal and diurnal variation of radon concentration indicates that the station is very rarely inside the surface inversion layer (Paatero *et al.* 1999). However, direct micrometeorological CO_2 flux measurements over a forest east of Sammaltunturi show that in summer





during some nights respiration from the forest ecosystem may increase CO_2 concentration at the fjeld top site, but during daytime the concentrations are well mixed (Aalto *et al.* 2002).

Carbon dioxide

 CO_2 has a long atmospheric lifetime and therefore the signal observed at the Pallas site can be influenced by long range transported marine and continental air, as well as anthropogenic sources (Aalto *et al.* 2002). Local influences are also possible due to boreal forest surrounding the fjeld where the site is located. However, the site is most of the time in touch with the free tropospheric air and the distance to major anthropogenic sources is long. Thus the possibility exists for measuring a representative CO_2 signal for continental background air. Representative measurements can be used for example for inversion modelling and detecting the global growth rate of CO_2 .

The Pallas CO₂ data set was analysed in order to select well-mixed air parcels free from dominating local influences and instrumental errors. Firstly, concentration recordings during station visits and equipment testing were removed. To accept an hourly CO₂ concentration, the mean wind speed was required to be higher than 2 m s⁻¹, the standard deviation of the hourly mean lower than 1 ppm and subsequent hourly values less than one ppm apart from each other. Daily medians were calculated from the selected data set. A harmonic function including a polynomial was fitted to the daily values, and standard deviation of the difference between the curve and hourly values was calculated for each day. Hourly data points with distance larger than three standard deviations from the curve were discarded. After these steps 62% of the original hourly values remained in the valid data set. Daily means were calculated from the new set requiring more than eight valid hours/day. Harmonic function and growth trend were obtained from the daily means according to methods pre-

Table 5. Meteorological parameters for the Sammaltunturi station, 1996–2002.

Parameter	Average	Min. daily average	Max. daily average	Min. hourly average	Max. hourly average	
Temperature (°C)	-1.1	-32.7	20.7	-33.4	24.6	
Wind speed (m s ⁻¹)	7.0	0.9	19.1	0	25.6	
Relative humidity (%)	84.9	28.6	100	16.3	100	
Pressure (hPa)	941.6	894.8	976.8	892.5	977.7	
Dewpoint (°C)	-3.8	-36.1	16.3	-36.9	17.8	
Global radiation (W m ⁻²)	118.3	0.1	370.0	0	892.8	
Visibility (km)	28.8	0.05	50	0.04	50	

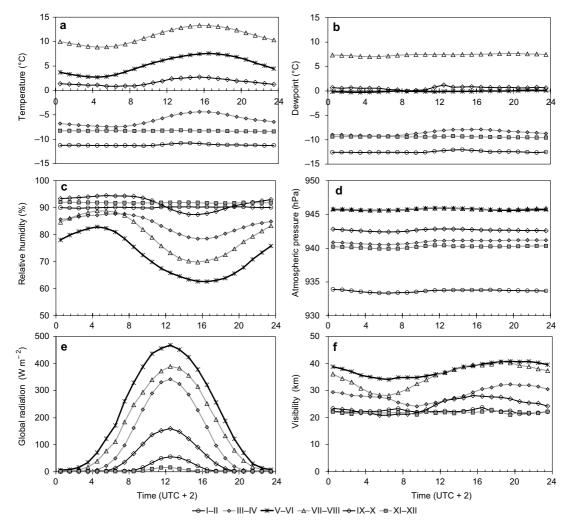


Fig. 3. Average bimonthly diurnal cycle of (a) temperature, (b) dewpoint, (c) relative humidity, (d) atmospheric pressure, (e) global radiation and (f) visibility at Sammaltunturi station, 1996–2002.

sented by Thoning *et al.* (1989). The calculation routine 'ccgvu' was utilised (available at http:// www.cmdl.noaa.gov/ccgg/resources/).

The fitting procedure was repeated for a data set where only measurements during 12:00-16:00 LT were utilised. According to boundary layer dynamics the measurement site is coupled with the surrounding forested terrain most probably during those hours. Both sets indicated similar average CO₂ growth rate of 1.9 ppm/year for the period Oct 1996–Oct 2002. However, the variation in growth rate was somewhat smaller when using the afternoon data, because lower concentration values were more frequent in the daytime. The growth rate indicated by the afternoon measurements is presented in Fig. 4. Due to missing data the growth rate does not fully catch the globally observed peak during 1997–1998 (Tans *et al.* 2002). Otherwise, the growth rate is in agreement with the global mean, and the daily means are rather smooth suggesting that the results are representative of large scale continental CO_2 concentration signal.

Ozone, NO, and sulphur dioxide

Ozone concentration in the boundary layer is a net effect of transport from the free troposphere, horizontal advection from other regions,

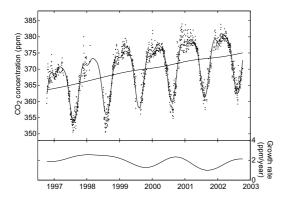


Fig 4. Carbon dioxide concentration at Pallas. Dots refer to daily means and lines to the growth rate, trend and a harmonic function fitted to the data.

deposition to the surface and net photochemical production including chemical sinks. All these factors affect diurnal cycle of ozone concentration (Fig. 5). If measurements are conducted at low-elevation sites with substantial vegetation, a nocturnal concentration minimum is usually observed when the wind speed is low (Rummukainen et al. 1996). Diurnal cycle is strengthened in polluted areas where photochemical formation produces a late afternoon maximum and titration by NO emissions a minimum during night. This is observed at Finnish monitoring sites in southern Finland such as Virolahti and Evo (Laurila 1996). At Sammaltunturi, average diurnal range is not observed in winter and even in summer it is very small, less than 5 ppb, which is typical for elevated and remote sites. Diurnal cycle of ozone concentrations has similarities to those of Radon-222 suggesting boundary layer mixing as one cause for variations, the other being photochemistry.

At high northern latitudes, strong seasonal variation of solar radiation divides the year into four periods regarding ozone formation. In winter, ozone precursors accumulate in the troposphere and act as a sink of ozone. In summer, they produce ozone, and the lifetime of ozone is shorter as photochemical processes are intensive. Spring and autumn are transitional periods when both directions of reactions alternate. These seasons are clearly shown, following analysis by Laurila (1999), as a relation between ozone and oxidation products of NO_x . As a surrogate of the

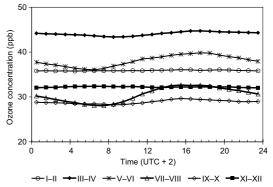


Fig. 5. Average bimonthly diurnal cycle of ozone concentration at Sammaltunturi, 1996–2001.

latter, total inorganic nitrate (gaseous HNO₂ and particulate nitrate) concentration is used, which is measured as 24-hour samples using combined particle and impregnated filters at the Matorova station. Total inorganic nitrate concentration indicates the amount of NO_x emissions that the air mass has received during its transport. Positive covariance between total inorganic nitrate and ozone concentration indicates photochemical ozone production, because in Scandinavia ozone production is controlled by NO_x concentration. The data is divided into four subgroups on a monthly basis according to the 10, 50, and 90 percentiles of total inorganic nitrate concentrations, and average ozone concentrations are calculated in these subgroups (Fig. 6).

The most unpolluted air masses are represented by the subgroup having the lowest total inorganic nitrate concentrations (percentile less than 10). In these clean air masses ozone concentrations are highest in March and April, with averages exceeding 40 ppb. The minimum is in July-August, when on average ozone concentration in unpolluted air mass is 25 ppb. In March-August, we observe photochemical ozone production because, on average, ozone concentrations are higher in the air masses which have higher total inorganic nitrate concentrations. In May-August the range of ozone concentrations between the highest and lowest total inorganic nitrate percentiles is 12-15 ppb and intermediate percentiles are between these highest and lowest percentiles. In winter a reversed order is observed as a result of missing photo-

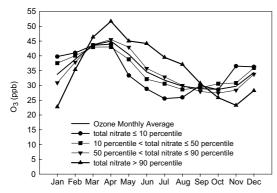


Fig. 6. Seasonal cycle of the relationship between ozone and total nitrate concentration in 1996–2001. For each month, daily average ozone concentrations are divided into four subgroups according to the total inorganic nitrate concentration 10, 50, and 90 percentiles. In each subgroup, mean ozone concentrations are plotted. In addition, the average monthly ozone concentration is shown.

chemical activity and air pollutants acting as a sink of ozone. The transition months are March, August and September. As compared with those at Utö island in the south-western archipelago of Finland, summer minimum and spring maximum are stronger at Pallas (Laurila 1999).

The most probable reasons behind the spring maximum of background ozone concentration are higher stratospheric flux due to seasonally high ozone concentrations in the lower stratosphere, long photochemical lifetime of ozone in winter, and low dry deposition velocity. Photochemical formation does not explain the growth of concentration before March, but after that it plays a role because of oxidation of the arctic winter reservoir of ozone precursors such as non-methane hydrocarbons (Laurila and Hakola 1996), and the correlation between ozone and total inorganic nitrate turns positive. According to the ozone soundings at Sodankylä, average concentrations in the lower free troposphere in spring and summer are equal and higher than in autumn and winter (Rummukainen et al. 1996). Within the boundary layer, ozone concentrations in summer are lower than in spring, which suggests higher dry deposition velocities to vegetation in summer as a cause of summer minimum.

Nitrogen oxide and nitrogen dioxide concentrations, based on measurements made with

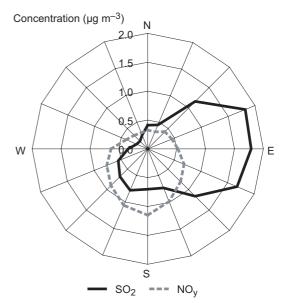


Fig. 7. Average sulphur dioxide (SO₂) concentration 1996–2001, and sum of all oxidised nitrogen species (NO_y) concentrations 2000–2001 in μ g m⁻³ classified by wind direction at Sammaltunturi.

Ecophysics instrument in 1998, are very low at this relatively remote site. At noon, average NO concentrations are 18 ± 11 ppt and 23 ± 11 ppt in May and June, respectively. Noontime calculated NO₂ concentrations are 25 ± 20 ppt and 61 \pm 48 ppt in May and June, respectively. During these measurements air masses were advected to Pallas from the Norwegian and Barents Seas and from northern continental regions where NO_x emission densities are very low. Thus, these NO concentrations are in the lower limit of summer concentrations in the northern boreal region. Under these low NO₂ conditions photochemical ozone production is very low or absent. However, NO₂ concentrations are not low enough for photochemical ozone destruction. Over more southern boreal regions average NO_x concentrations are higher. Previous NO_x measurements at Utö (Laurila et al. 1993) have shown that the average background NO_x concentration is ca. 1 ppb in the southern parts of Finland.

Emissions of NO_x in northern Europe seem to slightly increase their concentrations at Pallas. A recent study on photochemical processes in the remote boreal and arctic regions of Canada show even lower NO and NO₂ concentrations (Wang *et al.* 2003). In the boundary layer, Feb-

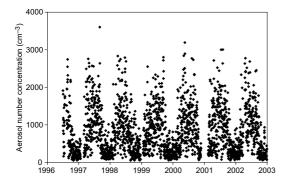


Fig. 8. Daily averages of total aerosol number concentration at Sammaltunturi, 1996–2002.

ruary–May monthly medians of NO and NO₂ concentrations varied between 1–9 and 8–10 ppt, respectively. The same study shows also that the most important species of total oxidised nitrogen species (NO_y) is PAN whose monthly median concentrations varied between 120–160 ppt in February–May. It is likely that the NO_x measurements by TEI42CTL reflect mostly PAN concentrations.

The mean value for sulphur dioxide concentration at Sammaltunturi station for the period 1996–2001 is 0.84 μ g m⁻³, which corresponds to 79% percentile. There is a very clear source area for the SO₂: Kola Peninsula in Russia (Tuovinen *et al.* 1993), ca. 450 km ESE of Pallas. This can be seen in Fig. 7, the highest SO₂ values are detected during easterly winds. Fig. 7 also depicts NO_y concentration as a function of wind direction. This shows that NO_y is transported from the south, where most of the Scandinavian and other European NO_y sources are located.

Aerosols

The average total aerosol number concentration at Sammaltunturi station is 800 cm⁻³, which is about half of what is observed in southern Finland (Komppula *et al.* 2003). In Fig. 8 daily averages at Pallas over six and a half years are presented. A clear seasonal variation can be seen, high particle number concentration values are found in spring and summer and lower values during winter. In spring and summer the daily averages may rise over 3000 cm⁻³ and

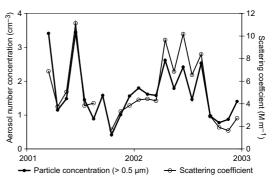


Fig. 9. Monthly averages of particle (larger than 0.5 μ m) concentration and total scattering coefficient at wavelength 550 nm.

in winter the daily averages may drop below 100 cm^{-3} .

The average of total scattering and backscattering coefficients of aerosol particles measured with an integrating nephelometer at wavelength 550 nm is 5.3 M m⁻¹ and 0.7 M m⁻¹, respectively. In Fig. 9 comparison of monthly averages of particle concentration (larger than 0.5 μ m) and total scattering coefficient over a period of almost two years is presented. The scattering coefficient correlates well with the particle concentration, as expected. Seasonal variation is also observed in scattering coefficient. High values are found in late spring and summer, and lowest values in autumn.

The aerosol particle concentration, size distribution and scattering coefficients depend on the origin of the air masses and secondary aerosol particle formation events. The low total aerosol particle scattering coefficients and number concentration in fall are due to prevailing westerly winds, indicating clean air masses from the North Atlantic. During summer prevailing easterly winds indicate air masses from the Kola Peninsula. This can be seen in Figures 8 and 9 as higher number concentration and scattering coefficient values. Aerosol particle formation events have a strong influence, especially to number concentration and size distribution, in spring (Komppula *et al.* 2003).

Radon

Usually the activity concentration of ²²²Rn is lower at Sammaltunturi than at Matorova due to

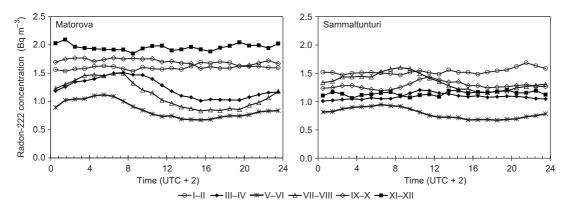


Fig 10. Average bimonthly diurnal variation of ²²²Rn concentration at Matorova and Sammaltunturi stations.

the higher altitude of Sammaltunturi (Fig. 10). The diurnal variation of ²²²Rn concentration during different seasons is rather similar at both sites. Minimum concentrations are found in May–June, when the radon exhalation rate is at its minimum due to the wet snow cover and high water content of the surface soil, and the vertical mixing of the boundary layer is efficient due to the long daylight duration.

At Matorova in July and August the diurnal variation is at its maximum due to the simultaneous strong radon exhalation and frequent nocturnal surface inversions. In September and October the diurnal variation decreases due to the decreasing amount of solar radiation causing vertical mixing. In November and December the diurnal variation is almost non-existent and the concentration is at its maximum due to the stable conditions in the troposphere owing to the polar night. Concentration in January and February is lower than in the previous two months due to the thickening snow cover which decreases the radon exhalation rate. In March and April the concentrations continue to decrease with increasing amount of snow. On the other hand, the diurnal variation starts to increase as the amount of solar radiation increases causing vertical mixing of the boundary layer air.

At Sammaltunturi in July and August the concentration maximum is reached a couple of hours later than at Matorova. The surface inversions block the radon to lower altitudes during night. After sunrise the warming of the groundlevel air breaks the inversion layer and releases the radon to the Sammaltunturi altitudes. This can be seen also in September and October when the diurnal maximum occurs a few hours later owing to the later sun rise and weaker solar radiation. As in Matorova, between November and February the diurnal variation is almost nonexistent and the concentration is at its maximum. The highest concentrations occur in January and February instead of November and December, as in Matorova.

Volatile organic compounds

The light hydrocarbons (C2–C6) play an important role in the tropospheric chemistry as precursors of CO, PAN (peroxyacetylnitrate) and ozone. Ozone formation depends on both hydrocarbon and NO_x concentrations, as well as the reactivity of the VOCs with hydroxyl (OH) radical. Generally, ozone formation is considered VOC sensitive close to emission sources, and as the air mass ages, the ozone formation becomes NO_x sensitive (Sillman 1999). In order to understand the oxidant formation at a rural location and to verify the emission inventories, the detailed VOC concentration measurements are necessary.

The VOCs are removed from the atmosphere mainly by a reaction with hydroxyl radicals, which are produced in sunlight. In winter, the hydroxyl radical concentrations are extremely low and therefore also the atmospheric lifetimes of VOCs are much longer resulting in concentration maximum. For example, the photochemical lifetime of butane at Pallas in January is about

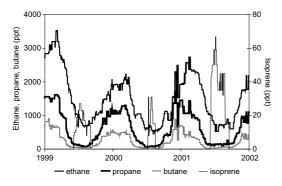


Fig. 11. 30-day running medians of ethane, propane, butane and isoprene concentrations at Sammaltunturi station, 1999–2001.

3.5 months and in July about 3.3 days. Figure 11 shows the annual cycle of some anthropogenic hydrocarbon (ethane, propane and butane) concentrations together with naturally produced isoprene concentration for the years 1999–2001. Isoprene is emitted from vegetation during the growing season and shows therefore in contrary to anthropogenic hydrocarbons a summer maximum. Concentrations of anthropogenic hydrocarbons begin to decrease in March when photochemistry and ozone formation starts (Fig. 6). Seasonal cycles of C2–C5 hydrocarbons at Pallas have been studied by Laurila and Hakola (1996).

Ethane is the most abundant VOC during all seasons, but in summer it comprises more than half of the total amount of light VOCs due to its slow reaction rate with hydroxyl radical (Fig. 12). The concentrations of the most reactive alkanes, such as pentane and hexane, are usually very low in summer at Pallas. Alkenes are more reactive than alkanes towards OH radicals, but in spite of this

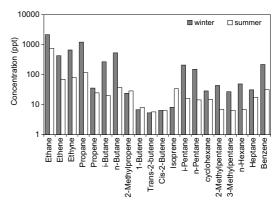


Fig. 12. Mean winter and summer concentrations of VOCs at Sammaltunturi station.

their concentrations are about the same throughout the year, except for ethene. This could implicate that some alkenes (propene, cis-2-butene, trans-2butene, 1-butene) have also biogenic sources.

Inorganic compounds and ions

Sulphur and nitrogen compounds

The range of the annual mean values of sulphur dioxide concentration from 1996 to 2001 is 0.43– 1.12 μ g m⁻³ at Matorova based on the laboratory analysis of impregnated filters. The annual mean concentration increased until 1998 and decreased to its lowest value in 2000. In 2001 the annual mean concentration increased again approaching the average value of 1996–2001 (Table 6). The highest annual mean concentration of sulphur dioxide in 1998 was due to an episode in

Table 6. Yearly averages of sulphate, sum of HNO_3 and NO_3^- as nitrogen, sum of NH_3 and NH_4^+ as nitrogen, and sulphur dioxide at Matorova station 1996–2001. In addition the last column shows yearly averages of sulphur dioxide concentration at Sammaltunturi station.

Year	SO ₄ ²⁻ (µg m ⁻³)	(HNO ₃ + NO ₃ ⁻)–N (µg m ⁻³)	$({ m NH_{_3}} + { m NH_{_4}}^+) - { m N} \ (\mu g \ m^{-3})$	SO ₂ (Mat) (µg m ⁻³)	SO ₂ (Sam) (µg m ⁻³)
1996	1.025	0.053	0.071	0.632	0.67
1997	1.023	0.045	0.110	0.746	1.00
1998	1.146	0.060	0.113	1.120	1.33
1999	1.451	0.072	0.129	0.848	0.87
2000	0.872	0.050	0.109	0.427	0.40
2001	0.956	0.059	0.117	0.712	0.76
Average	1.079	0.056	0.108	0.748	0.84

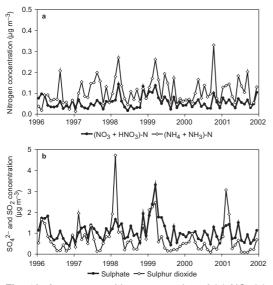


Fig. 13. Average monthly concentration of (**a**) NO₃⁻(p) + HNO₃(g) as nitrogen and NH₄⁺(p) + NH₃(g) as nitrogen and (**b**) sulphate and sulphur dioxide, Matorova, 1996–2001.

February. The monthly mean concentration was exceptionally high, 4.73 μ g m⁻³ (Fig. 13). The episode started on 6 February 1998 and lasted 11 days. The highest daily value was 17.5 μ g m⁻³. According to air mass back trajectories the air masses were coming from the Kola Peninsula. The monthly mean sulphur dioxide concentrations were highest in winter and spring from January to April every year, and the highest monthly mean concentration. In 2000 the highest monthly mean concentration was unusually low as compared with those in other years.

The annual mean concentration of sulphate in aerosols has not varied as much as sulphur dioxide concentration. The variation has been between 0.87 and 1.45 μ g m⁻³. The highest value was observed in 1999 and the lowest in 2000. The trend of the annual mean sulphate concentrations has been similar to the trend of sulphur dioxide. The annual mean sulphate concentrations increased until 1999 and the increase in the annual mean concentration in 2001 was smaller as compared with that of SO₂. The monthly mean sulphate concentrations were unusually high in February and in March 1999.

The annual mean concentration of the sum of gaseous nitric acid and particle bound nitrate $(HNO_3(g) + NO_3^{-}(p))$, and gaseous ammonia and particle bound ammonium ion $(NH_3(g) + NH_4^{+}(p))$ have varied to a lesser degree. Both sums have similar trends. The highest monthly mean concentrations of $HNO_3(g) + NO_3^{-}(p)$ were in March and November 1998 and in March 1999. The highest values were twofold the annual mean concentration. The monthly concentrations of $NH_3(g) + NH_4^{+}(p)$ have exceeded the double annual concentration limit more frequently.

Even though there were some high monthly mean concentrations at Pallas, the average concentrations, except for sulphur dioxide, were lower when compared with those at FMI's other stations in Finland (e.g. Oulanka, Ähtäri, Virolahti and Utö) in 1996–2001. Only at Ähtäri in southern continental Finland was the average concentration of sulphur dioxide slightly lower. The average concentrations of $HNO_3(g) + NO_3^-$ (p) were nearly equal at Pallas and Oulanka (290 km SE from Pallas).

Inorganic ions in precipitation

The annual volume-weighted mean concentrations of H⁺, Cl⁻, NO₃⁻, SO₄⁻²⁻, Mg²⁺, Ca²⁺, NH₄⁺, Na⁺ and K⁺ in precipitation are presented in Fig. 14. H⁺ concentration decreased to its lowest value in 1997 and remained low for the next three years. In 2000 it increased to a level as in 1995, but decreased again in 2001. The annual ammonium concentration had a significant increase in 1999. Nitrate concentration increased also to its highest value in the same year. Na+ and Cl- concentrations have decreased similarly since 1995. In 2000 and 2001 K⁺ concentrations have been half as low as compared with those in the previous years. The variations of Mg2+ and Ca2+ concentrations have been small. In 2001 Ca2+ concentration increased to its highest value. The trend of sulphate concentration has been quite similar to H⁺ concentration trend. The annual mean acidity of precipitation samples (pH values) has varied from 4.74 to 4.90. The lowest values were in 1995 and 2000 and the highest in 1996. The annual mean of electrical conductivity in precipitation has varied from 0.8 to 1 mS m⁻¹. The highest values were in 1995 and 2000 and the lowest in 1996–1998.

The annual amount of precipitation and deposition of H⁺, Cl⁻, NO₃⁻, SO₄²⁻, Mg²⁺, Ca²⁺, NH₄⁺, Na⁺, K⁺ during 1995–2001 are shown in Table 7. The highest annual depositions of H⁺, NO₃⁻, SO₄²⁻, Mg²⁺, Ca²⁺ and NH₄⁺ have been in 1995, 2000 and 2001. The higher values in 2000–2001 are partly due to higher precipitation amounts. The deposition trends have been very similar to the precipitation concentration trends. There has not been any large variation in Na⁺ and Cl⁻ depositions. K⁺ annual depositions have decreased considerably in 2000 and 2001 and Ca²⁺ annual deposition has increased since 1995. In 1999 the annual deposition of ammonium was nearly twofold the average value.

Average deposition values were generally lower during 1995–2001 compared with those at the other stations in Finland (e.g. Oulanka, Ähtäri, Virolahti and Utö). Only for Cl⁻, Na⁺ and K⁺ average values were slightly lower at Oulanka. General characteristic is very low concentrations of neutralising alkaline ions and ammonium. Nearly all of the chloride and respective anions originate from sea salt. Hydrogen ion concentration is roughly the same as in the southern and central parts of Finland despite the fact that sulphate and nitrate concentrations are one third of that observed in southern Finland.

Conclusions

Finnish Meteorological Institute chose Pallas as a site for a global GAW station because the area has no significant regional or local pollution sources. The National park, within which

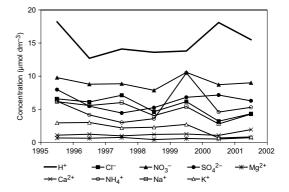


Fig. 14. Annual averages of main inorganic ion volumeweighted concentrations in precipitation, Matorova, 1996–2001.

Sammaltunturi station is situated, also ensures that the land use near the station is minimal (e.g. no forest cutting). At the same time the local topography allowed stations to be placed at different altitudes to get a better understanding of the structure of the lower troposphere of the area. The place has proven to be a good site for global background measurements, as Sammaltunturi station is very rarely inside the surface inversion layer. There are, however, episodes when the air masses are arriving from the Kola Peninsula. In these air masses especially sulphur dioxide concentrations can be tens of times higher than the average values.

The measurement programme will continue to expand at Pallas. In 2003 a GC system for measuring methane, dinitrogen oxide and sulphur hexafluoride will be installed at the Sammaltunturi station. CO_2 and radon gradient measurements will start in 2003–2004 at Kenttärova station.

Table 7. Yearly averages of precipitation amount and deposition in µmol m⁻² at Matorova station 1995–2001.

Year	Precipitation (mm)	H⁺	CI⁻	NO₃⁻	SO4 ²⁻	Mg ²⁺	Ca ²⁺	NH_{4}^{+}	Na⁺	K⁺
1995	454	8300	2990	4425	3617	288	499	2855	2784	1330
1996	405	5100	2454	3569	2214	247	499	1642	2262	1202
1997	321	4500	2285	2855	1434	247	324	928	1914	691
1998	494	6700	2285	3854	2588	206	599	1784	2001	1125
1999	456	6300	2793	4782	3087	247	574	4782	2479	1228
2000	576	10400	1834	4996	4116	288	599	2641	1609	384
2001	542	8400	2341	4854	3399	370	1048	2855	2305	435
Average	464	7100	2426	4191	2922	270	592	2498	2194	913

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