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Contaminants in the Atmosphere

AMAP- Nuuk, Westgreenland 2002-2004

NERI Technical Report, No. 547

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Datasheet

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Abstract: This report presents the results of atmospheric monitoring in Nuuk, Greenland. A long series of heavy metals and persistent organic Pollutants (POPs) have been measured and model calculations have been carried out supporting the interpretation of the results. Financially, the Danish Environmental Protection Agency supported this work with means from the MIKA/DANCEA funds for Environmental Support to the Arctic Region and the work is part of the Danish contribution to Arctic Monitoring and Assessment Programme, AMAP.

Keywords: West-Greenland, atmospheric long range transport, heavy metals, POPs, mercury, ozone

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Contents

Sammenfatning

Det er velkendt, at den grønlandske befolkning er udsat for tungmetaller og persistente organiske forureninger, POP'er, afhængig af, hvor stor en del af deres kost, der stammer fra marine dyr. Langtransport af luftforurening fra Canada og/eller USA og i episoder også fra Eurasia er antaget at være kilderne.

Formål

Formålet med målinger og modelberegninger i denne rapport er at bestemme tilførslen af udvalgte uorganisk og organiske luftforureningsforbindelser til Vestgrønland fra specielt Nordamerika.

Målesteder

Tungmetaller, inklusiv kviksølv samt, af O_3 (ozon), NOx (kvælstofmonoxid + kvælstofdioxid), og SOx (svovldioxid + sulfat) blev kontinuert målt på Lille Malene Bjerget tæt ved Nuuk, Grønlands hovedstad. Ligeledes blev der foretaget målinger af en serie POP'er på taget af ASIAQ's bygning i Nuuk. I denne rapport er resultaterne præsenteret og diskuteret. Fortolkninger af de målte data blev styrket ved brug af en receptormodel (COPREM) og en eulersk model (DEHM).

Tungmetaller

Der blev fundet lave niveauer af partikelbundne tungmetaller (partikelbundet kviksølv blev ikke målt) og SOx. En COPREM-analyse af data viste, at havsprøjt og jordstøv var de vigtigste kilder til den uorganisk partikelmasse, og at menneskeskabte kilder udgjorde mindre end 8% af denne.

Målte og modellerede niveauer af SOx og Pb

De målte niveauer af SOx og Pb (bly) i Nuuk er reproduceret med DEHM. Imidlertid gør de lave niveauer det umuligt at bruge målingerne til at forbedre og udvikle DEHM.

O_3 og GEM

I maj 2003 og i 2004 faldt gasfasekoncentrationen af kviksølv (GEM) fra baggrundsniveauet (ca. 1,5 ng m^{-3}) til 0,6 ng m^{-3}. I begge år var der i modsætning til Højarktis ingen korrelation mellem GEM og O_3. I 2003 skete faldet og den efterfølgende stigning langsomt, mens der var skarpe fald i koncentrationen i 2004. GEM faldet i foråret skyldtes højst sandsynligt langtransport af luftmasser, hvor GEM var omsat og deponeret.

Nye metoder

For første gang blev en passiv opsamler til måling af GEM testet og anvendt i felten. Et pyrolysesystem blev desuden anvendt sammen med en "TEKRAN 2537A mercury analyzer", hvilket gjorde det muligt at måle den samlede mængde kviksølv i atmosfæren og ikke alene GEM.

NOx

NOx målinger viste, at Lille Malene Stationen var påvirket af lokal, menneskeskabt forurening.

Klorpesticider

En serie chlorpesticider blev målt i lave koncentrationer. Stoffet med den højeste koncentration var hexachlorobenzen (HCB). Dog var opsamlingseffektiviteten af HCB meget dårlig på grund af dets store flygtighed. Koncentrationen af lindan (γ-HCH) i luften var højere i sommersæsonen (april-september) end resten af året, hvilket peger på episoder med langtransport af luftmasser fra det nordamerikanske kontinent, da lindan anvendes om sommeren.

PCDD/F

Polychlorinerede dibenzo-dioxiner og –furaner (PCDD/F'er) viste en udtalt sæsonvariation med høje koncentrationer om sommeren med maksimum i juli. Resten af året var niveauerne tæt på detektionsgrænsen. Sæsonvariationen skyldtes højst sandsynligt lokal affaldsforbrænding, men Nuuks forbrændingsanlæg kan udelukkes som kilde.

c-PCB'er og PCDF'er i biota

De lave koncentrationer af coplanare polychlorinerede biphenyler (c-PCB'er) i atmosfæren står i modsætning til observationerne i det grønlandske marine biota (alger, planter og dyr i havet) (Vorkamp *et al.*, 2004). Dette indikerer, at andre kilder end atmosfærisk langtransport er den primære kilde til c-PCB'er i Nuukområdet. I modsætning til dette er PCDF'erne dominerende i atmosfæren, men af lille betydning i biota. Disse eksempler viser, at det er vigtigt at forstå transporten af disse forbindelser mellem de forskellige matricer.

Nitro-fenoler

De små koncentrationer af nitro-fenol målt i sne indikerer, at de har en forsvindende lille effekt på den grønlandske vegetation.

Konklusion

Den overordnede konklusion er, at atmosfæren i Nuuk generelt er meget ren med undtagelse af små bidrag fra lokal forurening og bidrag fra langtransport af menneskeskabte forurening fra kilder i Nordamerika.

Anbefaling

Det generelt lave niveau af forurening (PCDD/F'er undtaget) i Nuuk og den konstaterede lille betydning af langtransport fra Nordamerika gør det klart, at overvågningssaktiviteterne i den grønlandske atmosfære skal fokusere på transporten fra Eurasia til Grønland. Derfor anbefales det at flytte målestationen fra Nuuk til Station Nord i Nordøstgrønland, men samtidig at revidere måleprogrammet, så det tager højde for transporten af de nye forureninger til Arktis og til strategien i de øvrige nationale programmer under AMAP.

Summary

The Greenlandic population is exposed to heavy metals and persistent organic compounds (POPs) due to the amount of their diet originating from sea animals. Long-range atmospheric transport of air pollution to western Greenland, possibly from Canada or USA and in episodes also from Eurasia, is suspected to be one of the main causes.

Aim

The Aim of the measurements and results of model calculations presented in this report was to determine the load of selected inorganic and organic pollutant to West Greenland from in particular North America.

Monitoring sites

At Lille Malene Mountain close to Nuuk, the capital of Greenland, continuous monitoring of heavy metals including mercury, as well as ozone, NOx and sulphate were carried out. Furthermore, a series of POP's were collected at the roof of ASIAQ building in Nuuk. The results are presented and discussed in this report. Use of a receptor model (COPREM) and an Eulerian model (DEHM) strengthened the interpretation of the monitoring data.

Heavy metals

The levels of particle bound heavy metals (excluding atmospheric mercury) and sulphur were very low. Analysis of data by COPREM showed that sea spray and crustal dust were the dominant source of inorganic particulate matter and that anthropogenic sources contributed with less than 8% of the inorganic particle mass.

Measured and modelled levels of Pb and SOx

The measured levels of SOx and Pb in Nuuk were reproduced by DEHM. However, the very low measured values of SOx and Pb concentrations made it impossible to use them to improve and develop the performance of DEHM.

O_3 and GEM

A strong depletion of GEM was observed in May 2003 with values down to 0.6 ng m^{-3} without any correlation to ozone concentrations as observed at higher latitudes. In 2004 the situation was different, some relative sharp depletion episodes were observed in April with concentrations down to 0.6 ng m^{-3}. Again there was not observed any correlation between GEM and O_3. The GEM depletion was in both years attributed to long-range transport.

New methods

For the first time a diffusive sampler for measuring atmospheric mercury was tested and used in the field. A pyrolyser was tested on a TEKRAN 2537A mercury analyser, which made it possible to measure directly total atmospheric mercury in the atmosphere.

NOx

NOx measurements clearly showed that the Lille Malene Station was affected by local anthropogenic pollution.

Chlorinated pesticides

A series of chlorinated pesticides was measured and found, generally at low concentrations. Hexachlorobenzene (HCB) was the compound found at the highest concentration. However, low collection efficiency was observed for HCB due to its high volatility. Lindane (γ-HCH) atmospheric concentrations were found to increase in the summer period (from April to September). This indicates a contribu-

tion of long-range transport from the North American continent, where this pesticide is still in use.

PCDD/F

PCDD/Fs show a pronounced seasonal variation, characterised by high concentrations in the summer with maximum in July. Otherwise, the concentrations were close to the detection limit. This annual profile was attributed to local sources and most probably from garbage burning, whereas the local municipal waste incinerator could be excluded as a possible source.

c-PCBs and PCDFs in biota

The low concentration of c-PCBs in the atmosphere is in contrast with observations in Greenlandic marine biota (Vorkamp *et al.*, 2004) . This indicates that other sources than atmospheric long-range transport is the primary c-PCBs source to the Nuuk area. Contrary to this, PCDFs dominate in the atmosphere but are insignificant in biota. These examples also highlight the need to understand the transport of these species between the various matrixes.

Nitro-phenols

The concentrations measured in snow indicate that deposition of nitro-phenols will have negligible effects on the Greenlandic vegetation.

Conclusion

The overall conclusion is that the atmosphere in Nuuk is generally very clean apart from a small local pollution and a small contribution from long-range transport from anthropogenic sources located in North America.

Recommendation

The general low levels of pollutants (except PCDD/Fs) in Nuuk and the little importance of sources in North America make it obvious that the monitoring activities in the Greenlandic atmosphere have to focus on the transport from Eurasia to Greenland. Therefore, it is recommended to move the station to Station Nord in Northeast Greenland but at the same time to revise the monitoring programme so it will include the transport of new contaminants to the Arctic and so that it is co-ordinated with other national monitoring programmes under AMAP.

Eqikkaaneq

Kalaallit Nunaamiut imarmiunik qanoq nerisaqartiginerat apeqqutaalluni aatsitassanit oqimaatsunit uumassusilinneersunillu mingutsitsinernit nungutikkuminaatsunit, POP'init, eqqugaasartut ilisimaarineqarluarpoq. Silaannaq mingutsitaq Canadamiit aamma/imaluunniit USA'miit ungasissumut ingerlaartartoq ilaatigullu aamma Eurasiamit pisartut tamatumunnga aallaaviusutut pasineqarput.

Siunertaq

Uuttortaanertigut pissutsillu assilillugit pisuusaartitsilluni naatsorsuinertigut nalunaarusiami matumani saqqummiunneqartutigut siunertaavoq akuleruuttunik uumassusilinneersunik uumaatsuneersunillu silaannarmut mingutserutaasunik pingaartumik Amerikamit Avannarlermiit Kalaallit Nunaata Kitaanut isaasoqarnerata paasilluarneqarnissaa.

Uuttortaaffiusut

Nuup, Kalaallit Nunaanni illoqarfiit pingaarnersaata eqqannguani Quassussuarmi aatsitassanik oqimaatsunik, aamma kviksølvimik kiisalu O_3 (ozon)-imik, NOx (kvælstofilte + kvælstoftveilte)-mik, aamma SOx (svovltveilte + sulfat)-imik ingerlaavartumik uuttortaasoqartarpoq. Aamma POP'it tulleriiaat Asiaq'p Nuummi illutaata qaliata qaavani uuttortarneqartarput. Tamakkunani paasisat nalunaarusiami matumani saqqummiunneqarlutillu oqaluuserineqarput. Uuttortaanertigut paasisat qanoq isumaqarnersiorneqarnerat sakkortusarneqarpoq malussarniusiaq (COPREM) aamma Euler-ip periaasia malillugu periaaseq (DEHM) atorlugit.

Aatsitassat oqimaatsut

Aatsitassat oqimaatsut silaarnarmi mikisuaraaqqaniittut annertunngitsuinnaat (taamaallaat kviksølv silaannarmi mikisuaraaqqaniittoq eqqaassanngikkaanni, tamanna uuttorneqanngimmat) aamma SOx malugineqarput. COPREM-analyse atorlugu paasissutissanik misissueqqissaarnikkut paasineqarpoq immap serpalinnera nunallu pujoralaa tassaasut silaannarmi mikisuaraaqqat uummaatsunit mingutsitaanerannut aallaaviunerpaasut, aamma mingutsitsinerit inuit pilersitaat taaneqartut 8%-iannit minnerusumik mingutsitsisuusut.

SOx aamma Pb qaffasissusiisigut uuttorneqartut takutinneqartullu

SOx aamma Pb (aqerloq) qaffasissusiisigut Nuummi uuttoorneqartut DEHM atorlugu pilerseqqinneqarput. Taamali aannikitsigigamik DEHM pitsanngorsassallugu ineriartortissallugulu atorneqarsinnaanngillat.

O_3 aamma GEM

Kviksølvip gas-inngorluni akuissusia (GEM) tunuliaqutaasup qaffasissusianeersoq (1,5 ng m^{-3}) majimi 2003-mi 2004-milu 0,6 ng m^{-3} tikillugu appariarpoq. Ukiuni arlariinni taakkunani Issittup Avannaarpiarsuani pissutsinit allaasumik GEM aamma O_3 imminnut ataqatigiinngillat. 2003-mi appariarneq malitsigalugulu qaffariarneq arriitsuinnarmik pipput, 2004-mili akuissuseq tassangaannaq appariarluni. GEM-ip upernaakkut appariarneranut silaannaat ungasissumut ingerlaartut GEM-ip allanngortinneqarluni toqqortarineqarfii pissutaassagunarluinnarput.

Periaatsit nutaat

Ilatsiinnarluni katersuissut GEM-imik uuttortaanermi atorneqartartoq sumiiffimmi pernaammik misilittarneqarlunilu atorneqarpoq. Aamma olia kissallugu ikummatissioriaaseq, pyrolysesystem, "TEKRAN 2537A mercury analyzer"-imik peqateqartillugu atorneqar-

	poq, taamalu silaannarmi kviksølv iluunngaat, GEM kisiat pinnagu, uuttorneqarsinnaalerluni.
NOx	NOx –imik uuttortaanerit takutippaat Quassussuarmi uuttortaavik qanittumi mingutsitsinermit inuit pilersitaannit sunnersimaneqartoq.
Klorpesticide-t	Klorpesticide-t assigiinngitsut akuippallaanngitsut uuttortarneqarput. Sananeqaat akuinnerpaasoq tassaavoq hexachlorobenzen (HCB). Kisianni HCB-mik katersuinerup pissarsiviunera assut annikippoq tamatuma katersoruminaassusia pissutigalugu. Lindan-ip (γ-HCH) silaannarmi akuissusia ukiup sineranut naleqqiullugu aasaanerani (april-september) annertunerpaavoq, tamannalu Amerikap Avannarliup nunavittaanit silaannaat ungasissumut ingerlaartarneranik tikkuussivoq, lindan ukiup taamaalinerani atorneqartarmat.
PCDD/F	Polychlorinerede dibenzo-dioxiner aamma –furaner (PCDD/F'it) akuissusii ukiup qanoq ilinera apeqqutaalluni allanngorartorujussuupput aasakkut akuinnerpaasarlutik julimi annerpaanngortartumik. Ukiup sinnerani tamakku qaffasissusiat uuttorneqarsinnaasup killinganiittarpoq. Ukiup qanoq ilinera najoqqutaralugu allanngorarnermut qanittumi eqqakkanik ikuallaasarneq pissutaasimassasoq ilimanartorujussuuvoq, kisianni Nuup ikuallaasarfianinngaanniiunngitsoq.
c-PCB'iit aamma PCDFiit uumassusilinniittut	Silaannarmi coplanare polychlorinerede biphenylit (c-PCB'iit) Kalaallit Nunaanni immami uumassusilinni (quajaatit immamilu naasut uumasullu) siumugassaasunisut inngillat (Vorkamp *et al.*, 2004). Tamatuma pasinarsisippaa silaannakkut ungasissumut ingerlaartartunit allat Nuup qanittuani c-PCB'nut peqqutaanerusut. Tamatuma akerlianik PCDF'it silaannarmi siumugassaanerpaapput, kisianni uumassusilinni pingaarutaat annikitsuinnaalluni. Assersuutissat tamakku takutippaat naaqqaviit assigiinngitsut akornanni imminnut kattuttut ingerlaartarnerata paasinissaa pingaartuusoq.
Nitro-fenolit	Nitro-fenol apummi uuttugaq annikitsuaraannaammat tamatuma pasinarsisippaa tamakku Kalaallit Nunaanni naasunut annikitsuaraannannguamik sunniuteqartut.
Inerniliineq	Ataatsimut isiginnilluni inerniliussaq tassaavoq Nuummi silaannaq ataatsimut isigalugu assut minguitsoq, tamaani mingutsitsinikkut sunniuttartut annikitsunnguusut aamma mingutsitsissutit inunnit pilersitat Amerika Avannarlermit ungasissumut ingerlaartartut eqqaassanngikkaanni.
Inassut	Nuummi mingutsitsinerup ataatsimut isigalugu annikitsuinnaasup (PCDD/F'it kisiisa pinnagit) aamma Amerika Avannarlermiit ungasissumut ingerlaartartut sunniutaata annikitsuinnaasup erseqqissarpaa Kalaallit Nunaanni silaannap alaatsinaanneqarnerani Eurasiamiit Kalaallit Nunaannut ingerlaartartut ukkanneqarnerusariaqartut. Taamaattumik inassutigineqarpoq Nuummi uuttortaavik Tunup avannaani Station Nordimut nuunneqassasoq, tamannali peqatigalugu uuttortaanermut periaaseq nutarterneqassasoq Issittumut Avannarlermut mingutserutaasut nutaat ingerlaartarneri aamma AMAP'ip ataani nunani ataasiakkaani ingerlassat allat naatsorsuutigineqarsinnaalersillugit.

1 Introduction

The Greenlandic population is known to be exposed to heavy metals and persistent organic compounds (POPs) from the part of their diet coming from sea animals. Long-range atmospheric transport of air pollution to western Greenland, possibly from Canada and/or USA and in episodes also from Eurasia, is suspected to be the main source for these pollutants. The pollutants are believed to enter the environment and food chain dominantly via atmospheric deposition.

The Nuuk Station is located on the Lille Malene Mountain at 345 m above sea level and serves as an atmospheric monitoring station for the south-western part of Greenland. The experimental results from the station can be used for determination of air pollution levels and seasonal variations in the most populated part of Greenland. The results can also be used for identification and quantification of the source types and the effect of long-range transport from the North American continent, which influences the atmospheric pollution and deposition in the region.

Furthermore, source areas and atmospheric transport pathways are identified by model calculations and it was intended to use the data for validation and further development of the Danish Eulerian Hemispheric Model (DEHM). The present approach has already proved to be very successful and led to an improved understanding of air pollution origin, emissions, transport pathways and deposition (Christensen, 1997; Skov *et al.*, 2004; Christensen *et al.*, 2004; Heidam *et al.*, 1999; Heidam *et al.*, 2004).

The present report is part of a continuous work on Greenland and is part of the Danish contribution to "Arctic Monitoring and Assessment Programme" (AMAP). The work improves the possibility of authorities to address the pollution problems in an international context and take the necessary legislative initiatives to abate the pollution. The Aim of the measurements and results of model calculations presented in this report was to determine the load of selected inorganic and organic pollutant to West Greenland from in particular North America.

2 Experimental

2.1.1 Field Stations

Lille Malene Station is located at 345 m above sea level at 64.1°N and 51.4°W and it is thus a Subarctic station. The station is located at Lille Malene Mountain just outside Nuuk (in Danish Godthåb), the capital of Greenland with about 13,500 inhabitants, see Figure 2.1.

Figure 2.1 Map over Greenland with Nuuk (Godthåb) located at Southwest Greenland.

At the foot of the mountain, Nuuk airport is located with one runway. All airborne traffic to Nuuk comes this way, see Figure 2.2. The only ski resort in Greenland is located on this mountain and the ski lift ends next to the monitoring hut.

Most transport of equipment to the Lille Malene Station is carried out during the snow season in January and February. Outside this season, transport with a helicopter is the only possibility for heavy and fragile equipment, whereas lighter things can be carried in backpacks up to the site.

Figure 2.2 Lille Malene Station with a view over Nuuk Town, and the runway of the airport

POPs

POPs were sampled on the roof of the ASIAQ building in Nuuk town, see Figure 2.3.

2.1.2 Measurements

Filter pack

Filter pack samples were collected on a weekly basis, and were sent to NERI for analysis. The following inorganic species were measured: SO_2, SO_4^{2-}, NO_3^-, NH_4^+, and elements analysed by PIXE (Proton Induced X-ray Emission) including Ni, Cu, Zn, As, and Pb. The high number of measured elements (22) provides a large amount of data, which is suitable for receptor analysis. In 2003, sampling and analyses were difficult due to occurrence of frequent episodes of fog at the Lille Malene Mountain, as a consequence the particle filters became wet. Lamps were installed at the end of the year to reduce the problem by heating the filters slightly. However, most samples were saved and the samples have been analysed, quality controlled and stored in the NERI-ATMI database. The description of sampling and analytical methods are given elsewhere (Heidam *et al.*, 2004; Hansen *et al.*, 2005).

Receptor analysis

All receptor models, including factor analysis, are based on the assumption that the original receptor site concentrations can be adequately explained by a linear combination of contributions from various relevant sources. With the Constrained Physical Receptor Model (COPREM) (Wåhlin, 2003) the corresponding linear equation is solved by an iterative method by which the chi-square (χ^2) is minimised within the limits imposed by constraints. χ^2 is the total squared distance between the measurements and the model values, measured in units of the uncertainties. Built-in model constraints exclude non-physical solutions (negative components in the source profiles and negative source strengths), and additional constraints can be included to fix profile components in constant ratios, partly or entirely. An initial profile matrix is setup in which the source vectors have the main characteristics of known sources, and the additional constraints are set up to maintain these characteristics, and to prevent the profiles from mixing together during the iteration. In this way any a priori knowledge about the character of the sources can be used to achieve a polarised solution as elements can be allowed to enter only in certain source profiles. COPREM also takes into account uncertainties, which are of particular importance for measurements close to the detection limit.

COPREM input

The input profiles and the constraints are shown in Table 2.3. The constrained values are written with bold types. The ratios between the constrained values in a column are not allowed to vary during the iteration in which both sources strengths and source profiles are fitted to give the minimum χ^2 value. No values are allowed by COPREM to enter into the negative space during the iteration. The measured values of Cl and Br were given infinite uncertainties because these elements to some extent evaporate during the PIXE analysis. The initial soil and sea profiles are taken from tables of elemental abundances ('Igneous rocks' and 'Sea water', respectively) in (Kaye & Laby, 1959). The constraints on the three anthropogenic sources 'Combustion', 'Metal_1' and 'Metal_2' were chosen to represent in the best way the variability of NSS-S (non sea salt sulphur), Zn and Pb, respectively.

Table 2.3. The COPREM input profiles and the constraints. The constrained values are written with bold types, the non-constrained values are written in italic. The ratios between the constrained values in a column are not allowed to vary during the iteration. The numbers are relative mass contributions within each source profile.

	Soil	Sea	Combustion	Metal 1	Metal 2
Al	*813*	**0.002**	*0*	*0*	*0*
Si	**2772**	**2**	*0*	*0*	*0*
S	**5.2**	**900**	**1**	**0**	**0**
Cl	**4.8**	**18900**	**0**	**0**	**0**
K	*259*	**370**	*0*	*0*	*0*
Ca	*363*	**402**	*0*	*0*	*0*
Ti	*44*	**0.001**	*0*	*0*	*0*
V	*1.5*	**0.0024**	*0*	*0*	*0*
Cr	*2*	**0**	*0*	*0*	*0*
Mn	*10*	**0**	*0*	*0*	*0*
Fe	*500*	**0.002**	*0*	*0*	*0*
Ni	**1**	**0.0017**	*0*	*0*	*0*
Cu	**0.7**	**0**	*0*	*0*	*0*
Zn	**0.8**	**0**	**0**	**1**	**0**
Ga	**0.15**	**0**	*0*	*0*	*0*
As	**0.05**	**0**	*0*	*0*	*0*
Se	**0.0009**	**0**	*0*	*0*	*0*
Br	**0.025**	**65**	**0**	**0**	**0**
Rb	**3.1**	**0.12**	*0*	*0*	*0*
Sr	**1.5**	**8**	*0*	*0*	*0*
Zr	*2.2*	**0**	*0*	*0*	*0*
Pb	**0.16**	**0**	**0**	**0**	**1**

Monitors

O_3, Hg, and NOx were measured continuously throughout the year at the station using automatic monitors. Monitor data from Nuuk are available on-line on the homepage of NERI, at "http://www2.dmu.dk/atmosphericenvironment/byer/forside.htm". The methodology and uncertainty have been presented previously (Heidam *et al.*, 2004; Skov *et al.*, 1997; Skov *et al.*, 2004). All monitors were sampling from the same sampling line except the one for GEM analysis. Unfortunately the central sample manifold was open from September 30 and the rest of the year 2004 and the error was found during the normal service at the station. Therefore all NOx and O_3 measurements were discarded in the quality control. Furthermore the sea breeze and general high frequency with foggy conditions made it difficult to measure GEM (Skov *et al.*, 2003). For this reason a soda-lime trap was installed to remove sea spray and acids. However these data and the results of inter-comparisons carried out in Barrow clearly showed that this might lead to a reduction of the GEM signal and thus lead to an underestimation of the GEM concentrations. Data has been discarded where we have evidence for this has happened.

POP's

In 2003 the equipment for POP measurements was constructed and tested. The system was ready at the end of 2003 and the collection was started at the beginning of January 2004, so the first full year data of the selected POPs can now be presented.

The POP sampling system designed and constructed by NERI was placed on the roof of the ASIAQ building in Nuuk town because of insufficient electrical power at Lille Malene to run a high volume sampler. Furthermore fog episodes were expected to interfere the sampling at the mountain site, see Figure 2.3.

Figure 2.3 Left picture: The inlet to the high volume sampler at the roof of ASIAQ, Nuuk and right picture the pump and the sample flow detection system

The construction and validation of the equipment were carried out at NERI in order to reduce the costs.

Selected POP's and nitrophenols

The POPs measured in the atmospheric comprised a series of chlorinated pesticides; α- and γ-hexachlorocyclohexane (HCH), Endosulfan, Heptachlor epoxide, Dieldrin, α and γ- Chlordane, Hexachlorobenzene, p,p′-DDT, p,p′-DDE, o,p′-DDT, o,p′-DDE, and Trifluralin. Oxychlordane is not included in the list as the compound turned out to have too high polarity to be analysed together with the other compounds. Furthermore polychlorinated dibenzo-p-dioxins (PCDDs) polychlorinated dibenzofurans (PCDFs) and coplanar PCBs (c-PCBs) are measured in the air samples. Finally, nitrophenols were measured in snow samples from Lille Malene Mountain.

Sampling site

The sampling in the town with more local pollution than at the Lille Malene site was not a big problem for the chlorinated pesticides as there are no sources of them in Greenland at all. Therefore local effects should be negligible for this group of compounds. On the other hand there are local sources of PCDDs, PCDFs and c-PCBs, which have to be taken into consideration in the interpretation of the measurements of these compounds.

Measurements of POPs in the atmosphere in Greenland have not been presented before; therefore the applied method is described below.

Sampling

Atmospheric samples of 3500-4500 m^3 were collected, during half monthly periods performed each month, from January to December 2004. The air sampling train consisted of a glass fibre filter (GFF) type A/E, 102 mm (Gelman), to collect particulate phase, followed by two cylindrical polyurethane foam (PUF) plugs (the company Søm og skumplastfabrik, Norway), 6 cm diameter and 5 cm long with a density of 0,02 g cm^{-3}, positioned in series. The flow was measured with a flow meter (rotameter) at the inlet of the sample train before and after each sample period. During sampling the flow was continuously measured using the pressure drop across an orifice. This pressure drop was registered and served to ensure that the flow was constant during sampling, by automatically adjusting the speed of the pump. In general the flow measured with the flow meter before and after sampling, was within 5% and the flow was set to about 250 litre min^{-1}. In a few cases this difference was much larger. The larger difference was observed for periods with very foggy and humid weather conditions and was coincident with the observation of wet filter and PUF plugs. The distribution of the various POPs between the first and second PUF plug remained the same during these wet meteorological conditions. The flow change could be reproduced using the two flow measurements and the registration of the pressure drop over the orifice. Therefore the sample volume under wet foggy conditions is not connected with additional uncertainty.

The uncertainty on the sample volume is estimated to be less than 10% (1 std. dev.). The uncertainty of the chemical analysis can be considered to be negligible. For the most volatile compounds (HCB and α-HCH) the main source of uncertainty is due to losses during sampling. Analysis of backup PUF had shown breakthrough of these compounds. For many samples, especially for those collected in the period June-September, the concentration of HCB on the second PUF was higher than the concentration on the first PUF. This means that a significant part is lost which can be more than 90% of the measured concentrations. For these samples the concentrations of HCB are not reported due to very poor collection efficiency. For the other samples the concentration of HCB and α-HCH collected on the second PUF has been used to calculate the collection efficiency. The concentration of the compounds on the first PUF has been corrected for the calculated collection efficiency. The acceptance criterion was that the concentration on the first PUF was higher than that on the second PUF. The calculated collection efficiency factor used to correct the concentrations of HCB and α-HCH on the first PUF varied between 6 and 80%. Thus, the uncertainty due to sampling efficiency below 100% has been minimised. However, the uncertainty of the calculated concentrations may be up to 50%. The breakthrough of α-HCH was strongly temperature-dependent, with low breakthrough percent (range: 4-9%) in the first part of the year and high breakthrough percent (more than 50% in the period June-August). In the following part of the year (September-December) the breakthrough percent gradually decreased from 70 to 11%. Also for this compound the concentrations measured in the summer period are underestimated.

List of POPs

The compounds included in the analytical program for POPs were α-HCH and γ-HCH (lindane), endosulfan, heptachlor epoxide, dieldrin, p,p′-DDT, p,p′-DDE, o,p′-DDT, o,p′-DDE, cis- and trans-chlordane, trifluralin; PCDDs: 2,3,7,8-tetrachlorodibenzodioxin (TeCDD), 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8- HpCDD and OCDD; PCDFs: 2,3,7,8-tetrachlorodibenzofuran (TeCDF), 1,2,3,7,8- PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8- HpCDF, 1,2,3,4,7,8,9- HpCDF and OCDF; c-PCBs: CB77, CB126 and CB169.

Analysis

For the analysis of chlorinated pesticides and trifluralin the samples were spiked with deuterium labelled γ-HCH, ^{13}C-HCB and ^{13}C-DDE and extracted by soxhlet with hexane/acetone (4:1, v/v) for 8 hours. The extract was evaporated, solvent exchanged to hexane and passed through a silica column (1 g). The analytes were eluted with 5 ml hexane followed by 5 ml hexane/dichloromethane (1:1, v/v). Both fractions were combined and evaporated to nearly dryness. The sample was reconstituted in isooctane and analysed by gas chromatography-high resolution mass spectrometry (GC-HRMS) at 10,000 resolution.

The analysis of PCDD/Fs and c-PCBs was done as described in Vikelsøe et al. (NERI Technical Report, in press, 2005) that follows a method adapted from European standard EN-1948 part 2 and 3 for analysis of PCDD/Fs in flue-gas. The samples were spiked with eleven $^{13}C_{12}$-labelled PCDD/Fs congeners and three c-PCBs congeners, and soxhlet extracted for 20 hours with toluene. Clean up was performed by column chromatography using SiO_2/NaOH, SiO_2/H_2SO_4, acidic Al_2O_3, active carbon (C AX-21, Supelco). The analysis of the extracts was performed by GC-HRMS. Field blank samples were analysed with each analytical series and the amounts of the analytes found in the blank sample were subtracted from the amount found in the air samples before the atmospheric concentrations were calculated.

Snow samples were analysed for the following nitrophenols: 4-nitrophenol, 2,4-dinitrophenol, 2,6-dinitrophenol, 2,6-dimethyl-4-nitrophenol, 3-methyl-2-nitrophenol, 3-methyl-4-nitro-phenol. Snow samples were extracted with solid phase extraction (SPE). Nitrophenols were analysed by liquid chromatography-tandem mass spectrometry (LC-MS-MS) following the method described elsewhere (Bossi *et al.*, 2002).

Toxic equivalents

The compounds in the PCDD/F family have very different individual toxicity. Therefore the results of those species are normally expressed in toxic equivalents, (TEQ). The toxicity of all congeners of a sample is set in relation to the most toxic compound, the "Seveso-dioxin" 2,3,7,8-TeCDD. In this way a series of many results can be expressed as a single number and the relative toxicity of 2,3,7,8-TeCDD is by definition set equal to 1. c-PCBs congeners exhibit a dioxin like toxicity, hence their toxicity are also expressed in relation to 2,3,7,8-TeCDD. WHO has made a system, which comprises all toxic equivalent factors (WHO-TEFs) for all toxic congeners of PCDD/Fs and c-PCBs. This system was used in the present study. Calculations of WHO-TEQ from concentrations of individual congeners in a sample were done multiplying the measured concentrations with the respective WHO-TEF.

DEHM

Finally computer simulations of the concentrations of sulphur species and Pb at Nuuk were carried out using the Danish Hemispheric Eulerian Model (DEHM). This model has been intensively used in AMAP and is at present used as the reference model. DEHM is developed especially to describe air pollution in the Arctic area and is described in detail elsewhere (Christensen, 1997; Christensen *et al.*, 2004; Heidam *et al.*, 2004; Skov *et al.*, 2004).

Global meteorological data

The model system is driven by global meteorological data obtained from the European Centre for Medium-range Weather Forecasts (ECMWF) on a 2.5° x 2.5° grid with a time resolution of 12 hours.

Grid resolution

The DEHM model is based on a set of coupled full three-dimensional advection-diffusion equations, one equation for each species. The horizontal mother domain of the model is defined on a regular 96x96 grid that covers most of the Northern Hemisphere with a grid resolution of 150 km × 150 km at 60°N. The vertical resolution is defined on an irregular grid with 20 layers up to about 15 km reflecting the structure of the atmosphere.

Chemistry module

The sulphur chemistry is simple linear, where the oxidation rate of SO_2 to SO_4^{2-} depends on the latitude and the time in the year, while for Pb there are no chemical transformations. The dry deposition velocities of the species are based on the resistance method. The wet deposition is parameterised with simple scavenging ratios for in-cloud and below-cloud scavenging. The removal rates for Pb are equal to the rates for SO_4^{2-}.

3 Results and discussion

3.1.1 Filterpack

In Figure 3.1 and 3.2 are summarised the results of compounds sampled by the filter pack in 2002 to 2004 at Nuuk. For all the compounds the concentration is very low.

Sulphur

In the case of sulphur compounds the very dominant compound is sulphate with very minor concentrations of sulphur dioxide. The highest levels of about 400 ng S m^{-3} are observed during late spring and early summer.

TNO_3 and ammonium

The maximum levels of ammonium and TNO_3 ($TNO_3 = NO_3^- + HNO_3$) of about 100 ng Nm^{-3} appear simultaneously in 2003 but a clear maximum did not appear for ammonium in 2004.

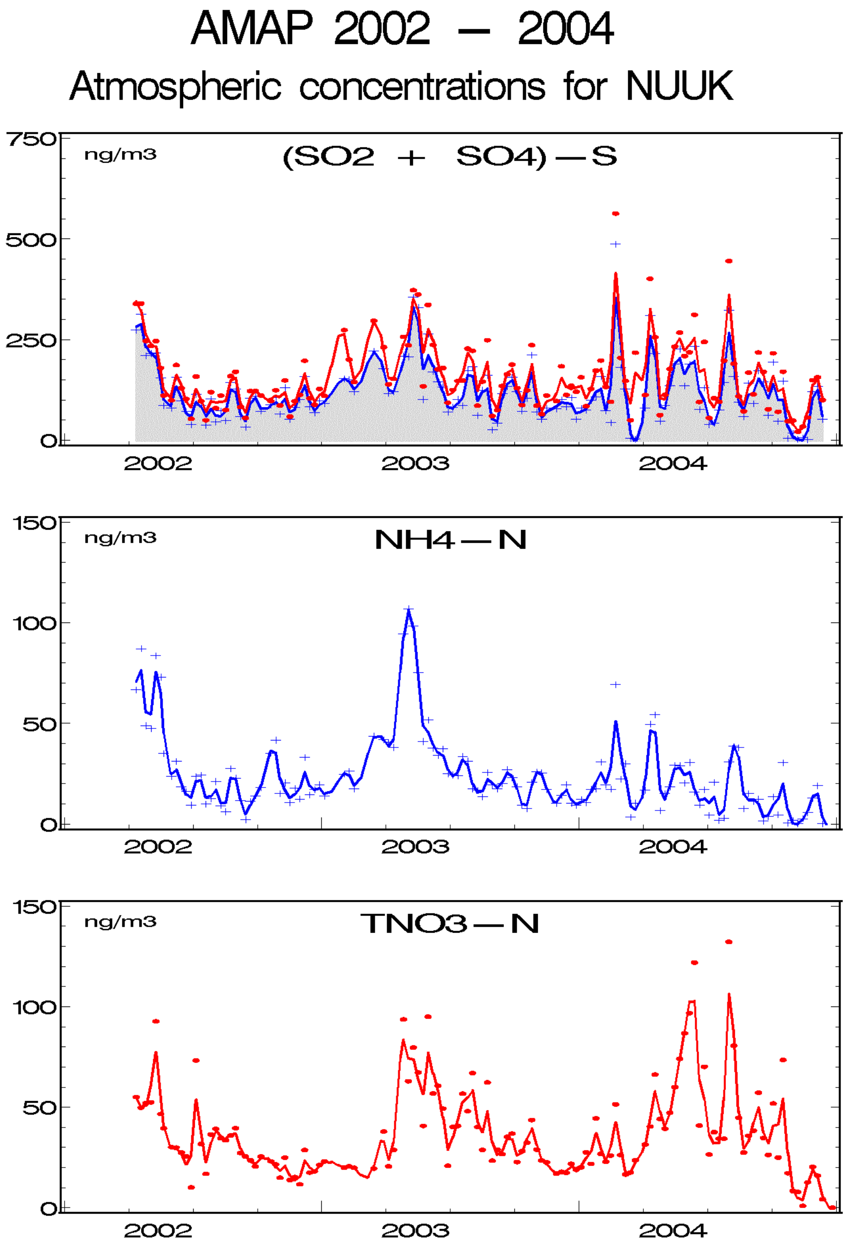

Figure 3.1 Filter pack measurements of SO_2, sulphate, ammonium and total nitrate, ($TNO_3 = NO_3^- + HNO_3$) at the Lille Malene Station, Nuuk, Greenland. The grey part of the SO_2 – SO_4^{--} graph is the SO_4^{--} and the upper line is the sum of the both sulphur compounds.

Elements

The concentrations of selected elements analysed by PIXE are shown in Figure 3.2 and average source contributions to each element (in ngm^{-3}) are listed in Table 3.1.

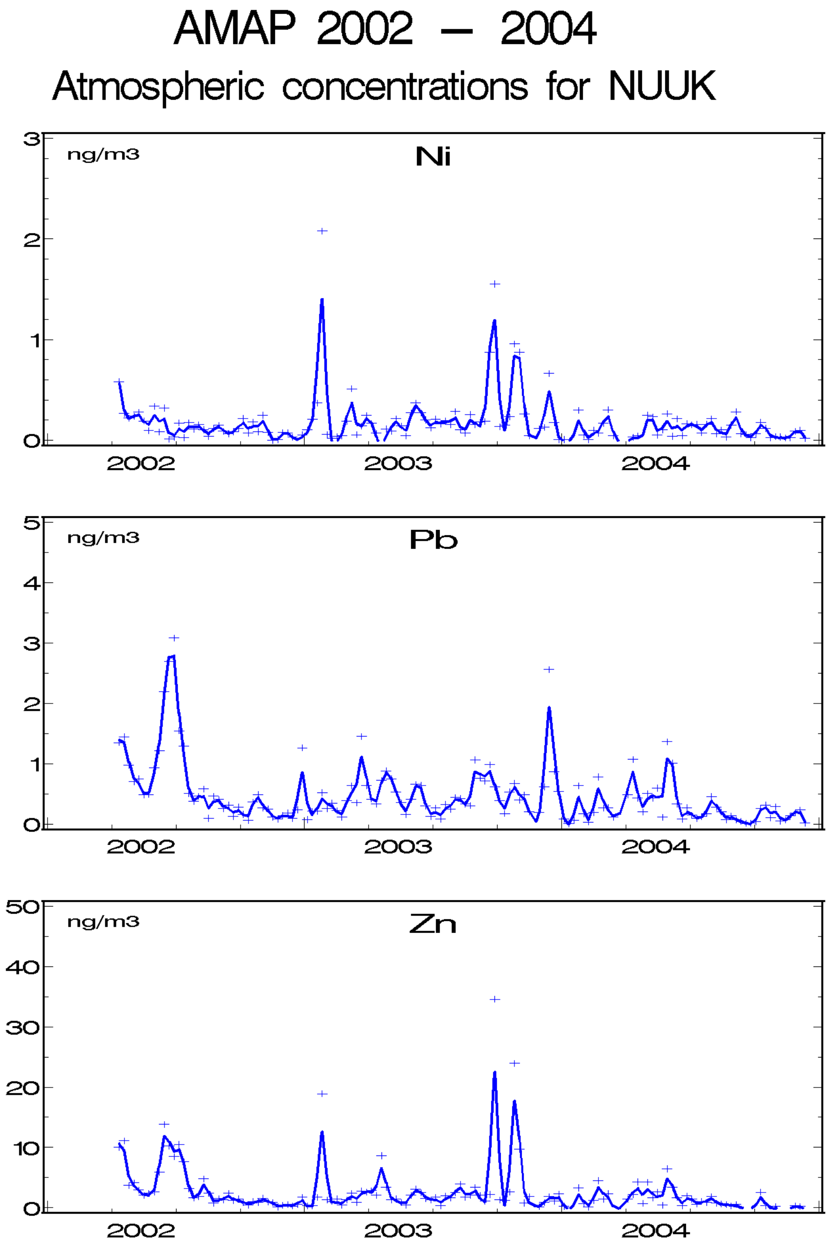

Figure 3.2 Time series of Ni, Pb and Zn at the Lille Malene Station, Nuuk Greenland.

The concentrations of the metals are in general low. The concentrations are in average slightly lower than the concentrations measured in north-eastern Greenland at Station Nord, where ‘arctic haze’ is observed in the winter (Heidam *et al.*, 2004). At Nuuk the pollution is distributed more evenly over the year (see Figure 3.2) than in north-eastern Greenland, where the levels are extremely low in the summer. The inorganic part of the particle mass (PM) is estimated to be on average 2.9 μgm^{-3} over the monitoring period. The estimate is based on calculations using the likely chemical composition involving non-PIXE elements and ions (as oxygen and carbonate). The corresponding mass correction factors are shown in the right part of Table 2.3. It is evident that most of the PM is due to sea spray. The second largest contributor is crustal material from soil erosion.

Comparison with Danish levels

The average PM (PM_{10}) at rural background sites in Denmark (Southern Scandinavia) is 22 μgm^{-3} where approximately half of it is inorganic material and thus it is about 3 times larger than the level observed in the present study.

Historical data

In 1979 to 1980 filter packs were operated at 5 places around Greenland (Heidam, 1984). Three stations were located at the westcoast of Greenland, Thule, Queqertarsuaq and Prins Christiansund, see Figure 1.1 (Prins Christiansund is not seen on the map but is located close to the most southern part of Greenland). Pb had a clear south north gradient with the largest concentrations at Thule at a geometrical mean of 1.82 ng m^{-3}, Queqertarsuaq at 1.28 ngm^{-3} and Prins Christiansund at 0.80 ng m^{-3}. For comparison the load at Station Nord was 4.98 ng m^{-3} from the same study. In Nuuk the geometrical mean of the data from August 2002 and to end of 2004 is only 0.48 ng m^{-3}.

The same comparison for sulphur does not show the same trend as Pb because of influence of sea salt sulphate at both Queqertarsuaq and Prins Christiansund.

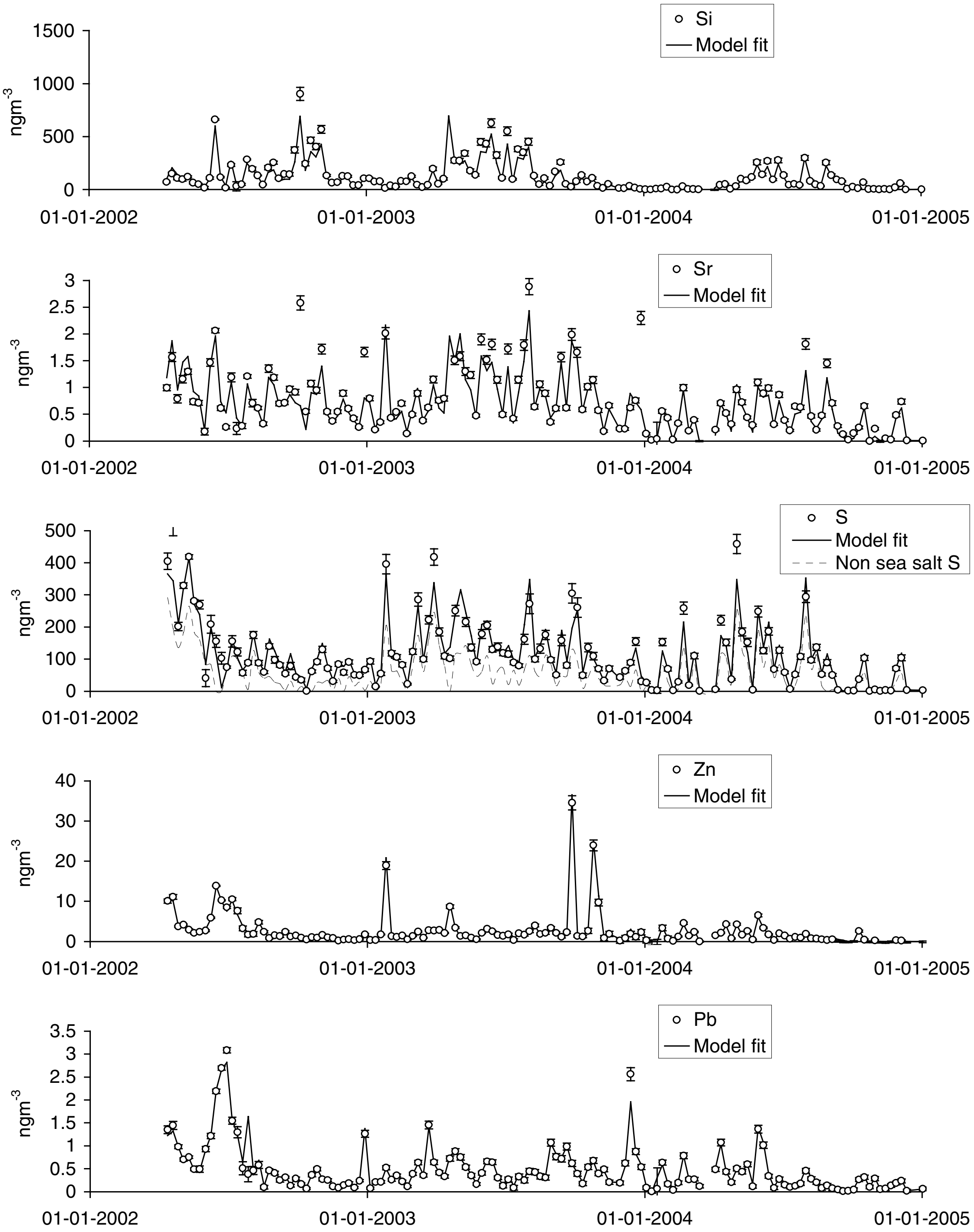

Figure 3.3. Time series and COPREM fits for some elements representing the major sources: Soil (Si), sea spray (Sr with only small contributions from other sources), combustion (S with a contribution also from sea spray), Zn and Pb (industrial sources).

Time series and model fit

The time series of some element concentrations that represent the major sources together with the model fit by COPREM are shown in Figure 3.3. COPREM can explain most of the concentration variability. The obtained source distribution is listed in Table 3.1.

Table 3.1. The resulting source profiles shown as the source contributions to the average concentrations in ngm^{-3}. The inorganic PM contributions (in ngm^{-3}) are calculated using the PM factors in the right part of the table. The organic PM contributions cannot be estimated using these data.

	Soil	Sea	Combustion	Metal 1	Metal 2	Sum	Measured average	Estimated chemical composition	PM factor
Al	39.29	0.00	0.00	0.00	0.36	39.65	44.36	Al_2O_3	1.89
Si	109.72	0.12	0.00	0.55	6.14	116.53	143.46	SiO_2	2.14
S	0.21	56.04	60.64	0.00	0.00	116.88	119.31	H_2SO_4	3.06
Cl	0.19	1176.91	0.00	0.00	0.00	1177.10	519.43	NaCl	1.65
K	9.98	23.04	0.67	5.02	1.39	40.10	42.89	K_2CO_3	1.77
Ca	15.96	25.03	3.65	4.66	0.00	49.30	51.66	$CaCO_3$	2.50
Ti	2.90	0.00	0.11	0.00	0.03	3.05	3.09	TiO_2	1.67
V	0.11	0.00	0.03	0.00	0.02	0.16	0.17	V_2O_5	1.79
Cr	0.08	0.00	0.01	0.02	0.00	0.11	0.11	Cr_2O_3	1.46
Mn	0.51	0.00	0.07	0.00	0.01	0.60	0.60	MnO_2	1.58
Fe	31.05	0.00	1.49	0.02	0.39	32.94	33.42	Fe_2O_3	1.43
Ni	0.04	0.00	0.04	0.06	0.00	0.14	0.17	NiO	1.27
Cu	0.03	0.00	0.10	0.11	0.02	0.26	0.36	CuO	1.25
Zn	0.03	0.00	0.00	2.71	0.00	2.74	2.62	ZnO	1.24
Ga	0.01	0.00	0.00	0.00	0.00	0.01	0.02	Ga_2O_3	1.34
As	0.00	0.00	0.02	0.00	0.00	0.02	0.02	As_2O_5	1.53
Se	0.00	0.00	0.03	0.00	0.01	0.04	0.05	SeO_2	1.41
Br	0.00	4.05	0.00	0.00	0.00	4.05	0.72	NaBr	1.29
Rb	0.12	0.01	0.00	0.00	0.00	0.13	0.11	Rb_2CO_3	1.35
Sr	0.06	0.50	0.00	0.09	0.05	0.70	0.79	$SrCO_3$	1.69
Zr	0.10	0.00	0.00	0.00	0.00	0.10	0.10	ZrO_2	1.35
Pb	0.01	0.00	0.00	0.00	0.49	0.49	0.48	PbO_2	1.15
PM	418.29	2221.13	198.50	25.50	17.61	2881.03			

Source distribution of sulphur

Approximately half of the sulphur at Nuuk is due to sea spray and the other half is attributed to combustion. Some of the sulphur attributed to the combustion sources may arise from natural emissions from the ocean from the photo-oxidation of dimethylsulphide and thus anthopogenic sources (combustion, Metal_1 and Metal_2) might be overestimated. Hence the contribution from anthropogenic sources at 8% of the inorganic particle mass represents an upper limit, see Table 3.1.

3.1.2 Model calculations

Traditionally we have focused on the description of SOx and Pb in the Arctic as important tracers for anthropogenic pollution and formation of Arctic haze (AMAP, 2003). Therefore they are also used here for comparison between model results and measured concentration. The measured and modeled concentrations of SOx (sum of SO_2 and SO_4^{2-}) are shown in Figure 3.4, and a similar comparison for Pb is shown in Figure 3.5.

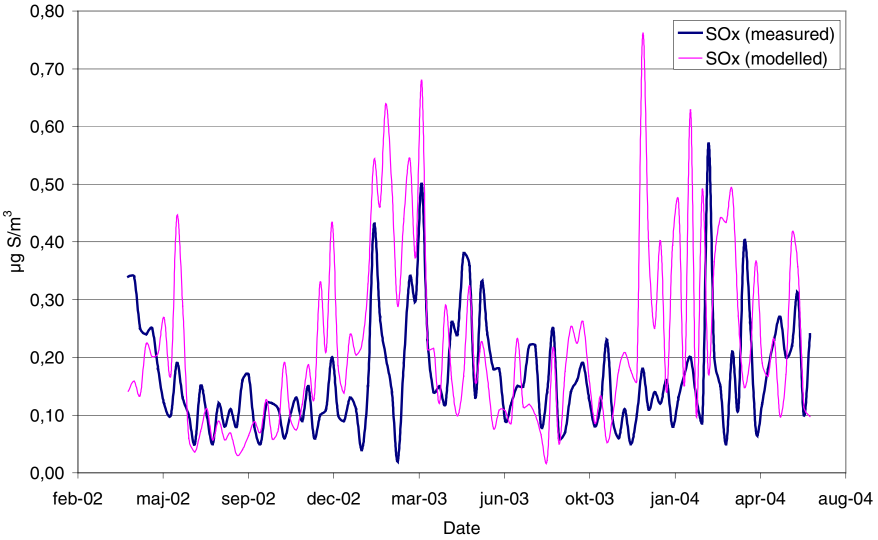

Figure 3.4 Comparison of model output and measured concentration of SOx (= $SO_2 + SO_4^{2-}$) in Nuuk

Average concentration

The levels of both modelled and measured values are very low. The average concentration for the period was respectively 0.17 µg S m^{-3} and 0.22 µg S m^{-3} for measured and modelled values, which is in excellent agreement. For lead the levels were also low. The respective average values were 2.46 ng m^{-3} and 1.17 ng m^{-3} for measured and modelled values, so that the measured concentrations are significant larger than the modelled ones.

Seasonal variations

In all the three years 2002, 2003 and 2004 DEHM predict a clear maximum in the SOx concentration during winter. This maximum is much weaker in the measured values. Contrary there is not any seasonal trend for lead. However, some transport episodes are identified where the measured values are much larger than the modelled values. From COPREM analysis we know that lead nearly exclusively can be assigned to metal production.

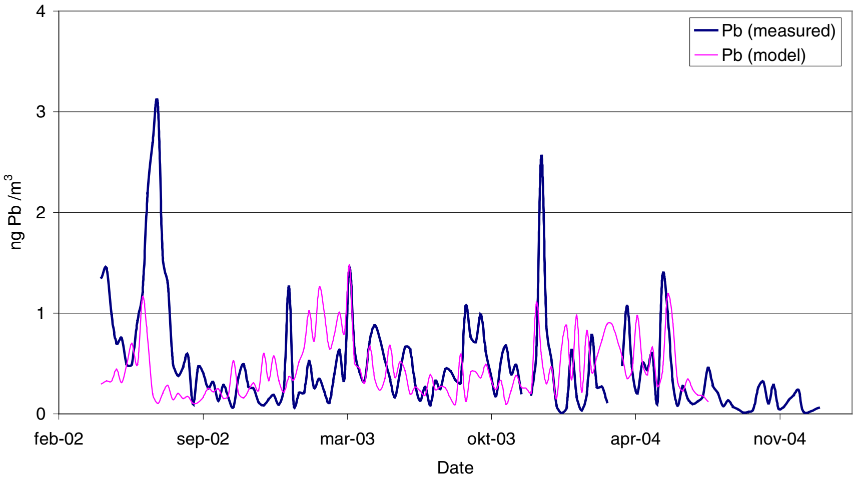

Figure 3.5 Comparison of model output and measured concentration of Pb in Nuuk.

The measured levels of SOx and Pb in Nuuk are reproduced by DEHM. But the very low values make it impossible to use the measured SOx and Pb concentrations to improve model performance of DEHM. The uncertainty on the measurements is too large and only very small variations in the emissions in the source regions mainly located in North America will change the concentrations significantly at Nuuk. This is in fact also the main reason for the discrepancy between measured and modelled seasonal pattern.

3.1.3 Monitor measurements

The concentrations of O_3, gaseous elemental mercury (GEM), NO and NO_2 are shown in Figures 3.6 to 3.7.

GEM and ozone

A strong depletion of GEM is observed in May 2003 with values down to 0.6 ng m^{-3} without any correlation to ozone concentrations as observed at higher latitudes (Schroeder *et al.*, 1998); (Lindberg *et al.*, 2002) (Berg *et al.*, 2003; Skov *et al.*, 2004) and the depletion is observed later than those in the High Arctic. In 2004 the situation was different; some relative sharp depletion episodes were observed in April with concentrations again down to 0.6 ngm^{-3}. Though the pattern looked much more like the one observed in the high Arctic there was not observed the same strong correlation between GEM and O_3 (Hansen *et al.*, 2005). These observations indicate that other mechanisms must be responsible for the GEM removal than the reaction with Br that most probably is the main removal channel at more northern locations (Goodsite *et al.*, 2004). In fact, filter pack measurements showed only very low concentrations of bromine at Nuuk, and contrary to the measurements at Station Nord the bromine was found to be only associated with particles. The levels of bromine at Station Nord were also much higher (Skov *et al.*, 2004).

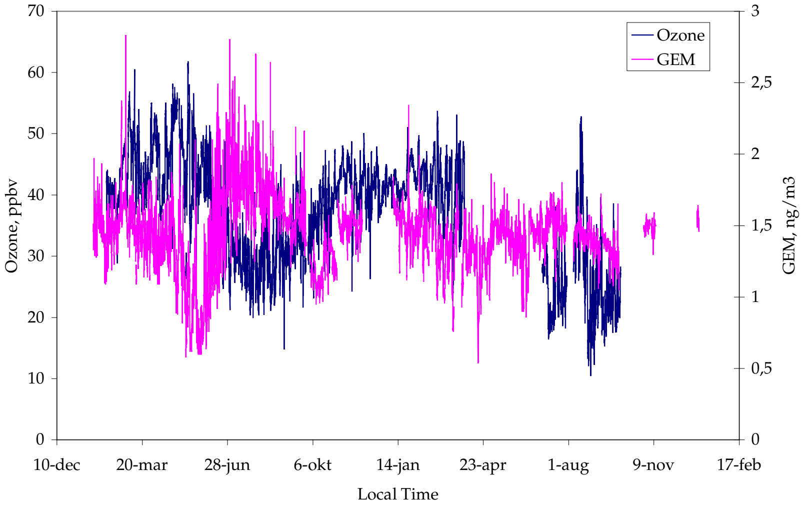

Figure 3.6 Ozone and Gaseous elemental mercury (GEM) at the Lille Malene Station in 2003 and 2004.

Hemispheric background levels

Outside the depletion period the concentration of GEM was close to 1.5 ng m^{-3} which is considered being the hemispheric background concentration. O_3 varied between about 50 ppbv and 25 ppbv with the maximum value in late winter and early spring and the minimum in July. These levels are also close to hemispheric background concentrations (Skov *et al.*, 1997).

Spring campaign, test of new methods

In the spring 2004 the measurements at Nuuk of GEM were extended with measurements using newly developed methods. Thus for the first time in the world diffusive samplers were used successfully to measure GEM in the atmosphere. The method was validated against the TEKRAN Hg monitor (Daugaard *et al.*, 2005). This method will make it possible in future to measure atmospheric GEM with high geographical resolution. Finally a pyrolyser was tested on a TEKRAN 2537A mercury analyser, which made it possible to measure total atmospheric mercury in the atmosphere (Hansen et al. 2005).

NOx

NO and NO_2 are good indicators for local pollution from combustion processes for example in the transport sector. In Figure 3.7 the concentrations of the two species are shown for 2003. There is a series of distinct peaks. Between 15 January and 27 April the peaks were much higher than the rest of the year.

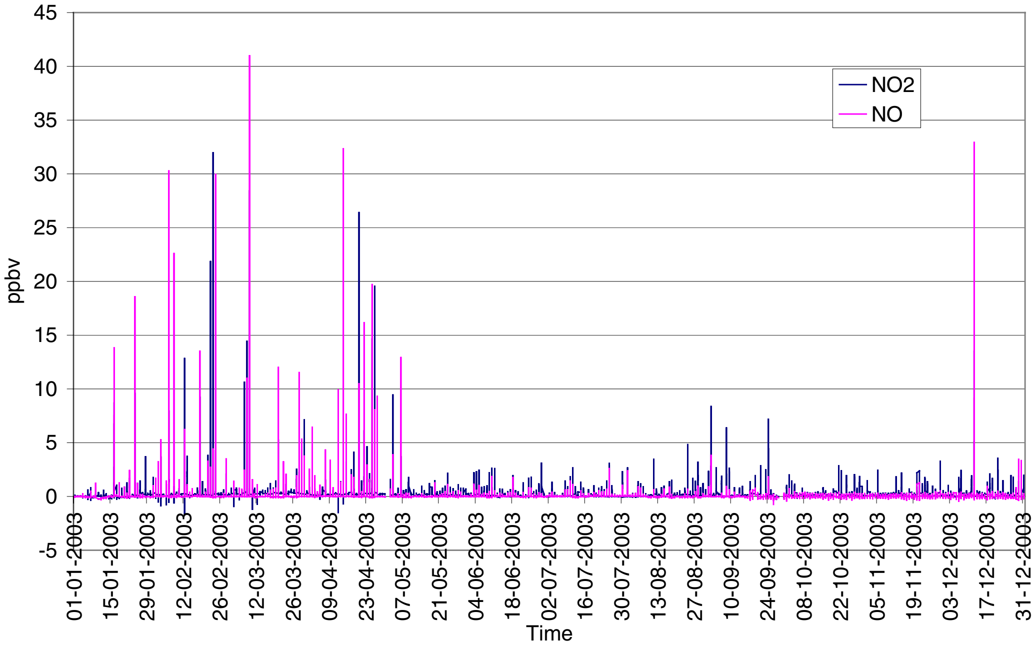

Figure 3.7 NO and NO_2 at Lille Malene, Nuuk in 2003

Local sources

The concentration has many sharp peaks that are strong evidence for the presence of very local sources. During the period from January to the end of April when the ski lift was open, snow scooters and "piston bulls" passed close to the monitoring hut causing high levels of NOx After the ski lift closed the levels of NOx decreased substantially and remained low until January next year when the ski lifts reopened.

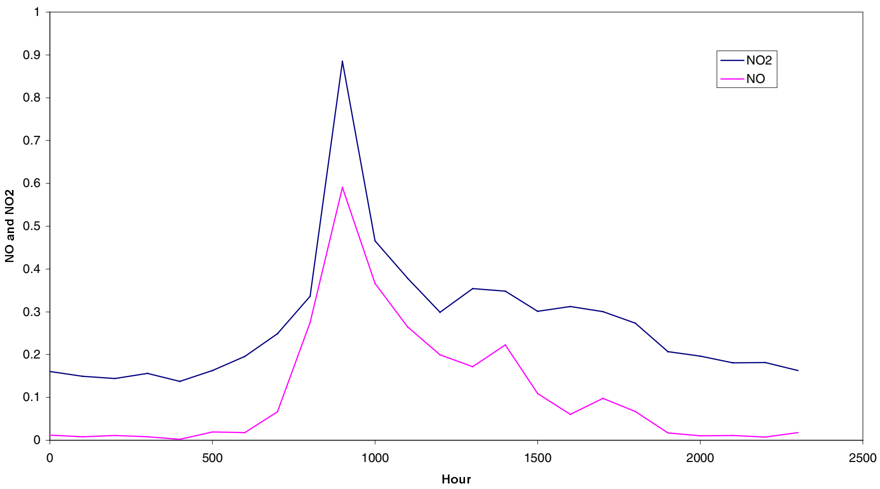

Figure 3.8 Average daily profile of NO_2 and NO at the Lille Malene Station obtained by averaging the concentrations from 2003 over a specific time of the day.

Daily profile

The data in Figure 3.7 was used to calculate an average daily profile, see Figure 3.8. A distinct peak was observed at 9:00 with a slow down slope reaching a minimum at night. This observation is in agreement with the above interpretation that the activities in fact started at 9:00 with the preparation of the ski slopes.

3.1.4 POP's

The results are summarised in Table 3.2. Concentrations of POPs in Nuuk can be compared to those measured in Eastern Canadian Arctic, since transport of air masses from the North American continent is the predominant source of these compounds. In the following paragraph we have compared the results from Nuuk with the data collected during the last 10 years at two stations in Eastern Canada, Alert (82° 28' N, 62° 18' W) and Kinngait (72°42'N, 77°59' W) (Hung *et al.*, 2005; Macdonald *et al.*, 2000; Halsall *et al.*, 1998).

HCB

HCB was the most predominant compound observed in the atmosphere, followed by α-HCH, endosulfan and γ-HCH (lindane). Emission of HCB to the atmosphere is mainly due to industrial activities. As discussed in section 2.1.2, the concentrations of HCB might be underestimated due to losses during sampling. The average concentration calculated for the months when the collection efficiency was considered satisfying was 128.1 pg m^{-3}, a value which is about twice as much as the average concentration in Kinngait (63.4 pg m^{-3}). Since the uncertainty of the HCB measurements is quite high (estimated to be 50%), the concentration measured at Nuuk is in fact not significantly different from those at Kinngait.

α and γ-HCH

The concentrations of most chlorinated pesticides did not show any seasonal variation (Figure 3.9), with the exception of γ-HCH (Figure 3.10), which had a maximum of 11.1 pg m^{-3} in August. Annual average concentrations of γ-HCH was 4.98 ± 2.54 pg m^{-3} These concentrations are lower than those measured in 1995 at Kinngait (9.38 pg m^{-3}). γ-HCH is still used in North America, particularly in the Canadian prairies for canola seed treatment. The air mass trajectories moving from the prairies to the Central Arctic latitudes have been found to contribute to the atmospheric load of contaminants by 30% (Waite et al., 2005). The lower concentration of γ-HCH measured in Nuuk respect to Kinngait may be due to deposition of this compound with precipitation during transport from North America to Greenland. γ-HCH is more water-soluble than α-HCH and therefore more easily scavenged by precipitation. The use of α-HCH is forbidden both in Europe and North America. Thus, the presence of α-HCH in the atmosphere is not determined by primary sources but is mostly the result of re-emission from terrestrial and aquatic surfaces previously contaminated by α-HCH. α-HCH concentrations in Kinngait in the period 2000-2001 have decreased by a factor 5 since 1995 (Hung *et al.*, 2005). α-HCH concentrations are expected to decrease, since the use of technical HCH (containing 55-80% α-HCH) has been banned by the two major users, China and the former USSR.

Endosulfan

Endosulfan average annual concentrations in Nuuk (6.32 ± 1.84 pg m^{-3}) are similar to those found in Kinngait in 1995 (5.64 pg m^{-3}). Atmospheric endosulfan concentrations are not expected to decrease, since this compound is a pesticide currently used in the North American continent.

Chlordanes

The use of chlordanes has been banned in most European and North American countries. The ratio α/γ-chlordane observed in Nuuk is about 1. The α/γ ratio in technical chlordane is 1.16. A lower ratio indicates a weathered chlordane profile, as the α isomer is more susceptible to degradation than the γ isomer. The sum of the concentrations of Chlordanes in Nuuk is 0.9 pg m^{-3}, which is similar to the average concentration in Kinngait in 1995 (1.39 pg m^{-3}).

Dieldrin

Dieldrin has been banned in most western industrialised countries. The atmospheric concentration in Nuuk (2.50 ± 1.02 pg m^{-3}) is similar to those found in Kinngait (1.45 pg m^{-3}) and Alert (1.18 pg m^{-3}) in the period 1994-1995.

Heptachlor epoxide

Heptachlor epoxide is a transformation product of the pesticide heptachlor. The major source of heptachlor epoxide in the atmosphere is the metabolism of heptachlor in soils followed by evaporation (Bidleman *et al.*, 1998). The annual average concentration in Nuuk (0.75 ± 0.31 pg m^{-3}) is close to that measured in Kinngait (1.08 pg m^{-3}).

DDT and DDE

Although DDT has been banned or severely restricted in many countries, it is still used in others, especially in Asia and in the tropical regions. The parent compound DDT was not found in any sample from Nuuk, whereas the degradation products p,p′-DDE and o,p′-DDE were found in most samples with annual average concentrations of 0.38 ± 0.19 and 1.14 ± pg m^{-3}, respectively. A clear seasonal variation was observed for o,p′-DDE with significantly higher concentrations in the period August-September. This variation may be due to changed emissions (e.g. from previously contaminated soils) as a function of temperature.

Trifluralin

Trifluralin is not a persistent compound but it has been included in the monitoring program as a currently used pesticide, since its extended use in the North American continent and in Europe, together with its relatively high vapour pressure (6.1 mPa) makes it a good candidate for atmospheric transport. Trifluralin has been previously measured in the atmosphere at Alert at average concentration of 0.12 pg m^{-3}. Trifluralin was only sporadically found in samples from Nuuk collected in winter and autumn and never during the summer season with an annual average concentration of 0.24 ± 0.22 pg m^{-3}.

Table 3.2. Annual mean and median concentrations of chlorinated pesticides and trifluralin (pg m^{-3}), as well as PCDDs, PCDFs and c-PCBs (fg m^{-3} WHO-TEQ) in Nuuk together with minimum and maximum values. The d/n column indicates number of detected concentrations above detection limit.

Compound	d/n	Mean	Median	Min	Max
alpha-Chlordane	13/13	0.4	0.4	0.2	0.9
gamma-Chlordane	13/13	0.5	0.5	0.3	0.7
4,4’-DDT	1/13	0.2	0.2	n.d.	0.2
2,4-DDT	0/13	n.d.	n.d.	n.d.	n.d.
p,p’-DDE	13/13	0.4	0.3	0.2	0.8
o,p’-DDE	11/13	1.1	0.2	0.0	5.0
Dieldrin	7/13	2.5	2.6	0.9	3.6
Endosulfan I + II	13/13	6.3	5.9	3.7	10.3
Heptachlor epoxide	13/13	0.8	0.7	0.3	1.2
Hexachlorobenzene	13/13	128.1	*	*	*
α-HCH	13/13	18.3	*	*	*
γ-HCH (Lindane)	13/13	5.0	4.6	2.2	11.1
Trifluralin	4/13	0.2	0.2	0.0	0.6
PCDDs	3/6	6.7	4.2	0.3	20.2
PCDFs	4/6	12.4	10.5	0.5	31.5
c-PCBs	3/4	0.6	0.6	0.2	1.0

*Not calculated due to missing results in the summer period (poor collection efficiency)

The seasonal variation of the most abundant chlorinated pesticides is shown in Figure 3.9 and the least abundant chlorinated pesticides are shown in Figure 3.10.

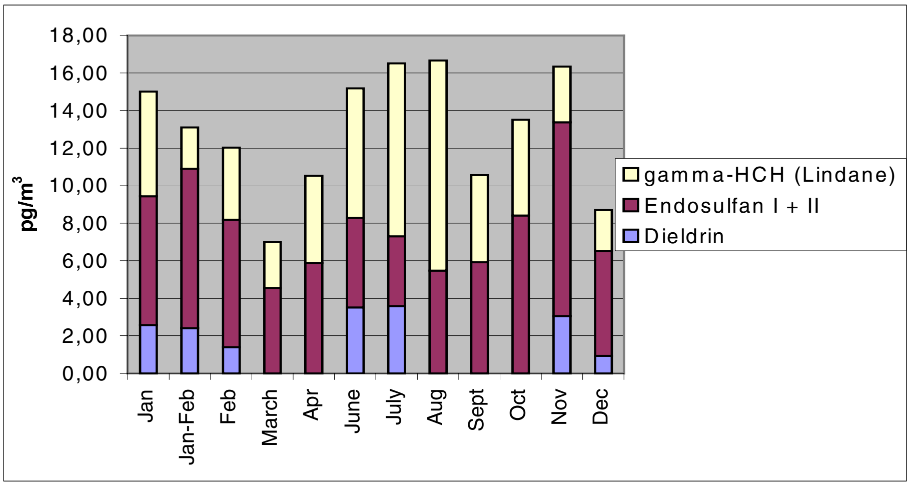

Figure 3.9 Concentrations of most abundant chlorinated pesticides in the air in Nuuk 2004 with half-monthly sampling periods carried out each month.

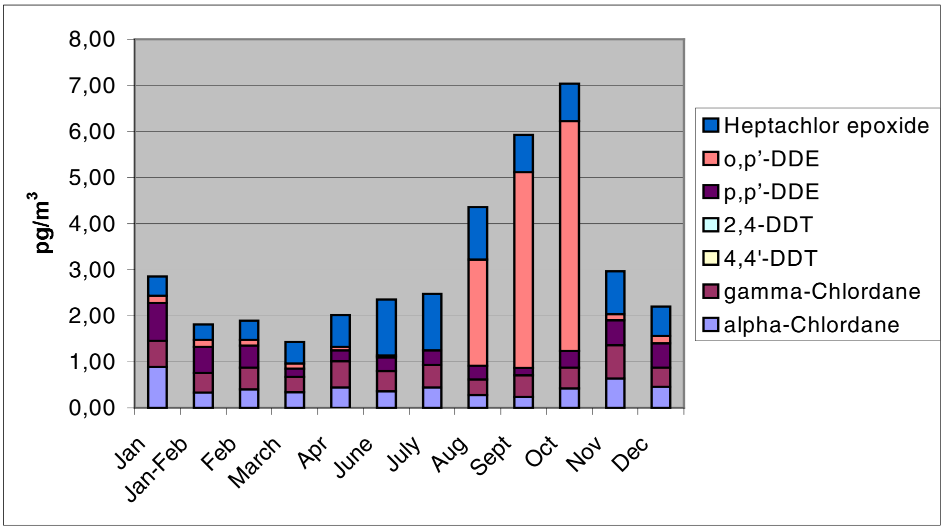

Figure 3.10 Concentrations of chlorinated pesticides of least abundant compounds in air in Nuuk 2004 with half-monthly sampling periods carried out each month. (Trifluralin has been left out).

PCDD/Fs

In the atmosphere PCDFs were found in much higher concentrations than c-PCBs. This contrasts with what is found in marine Greenlandic biota, where c-PCBs are dominating (Vorkamp *et al.*, 2004). This observation indicates the existence of a complicated mechanism for the transport between abiotic and biotic matrices.

The sum of C-PCB , PCDF and PCDD, respectively are shown in Figure 3.11 in the unit fgm^{-3} WHO-TEQ.

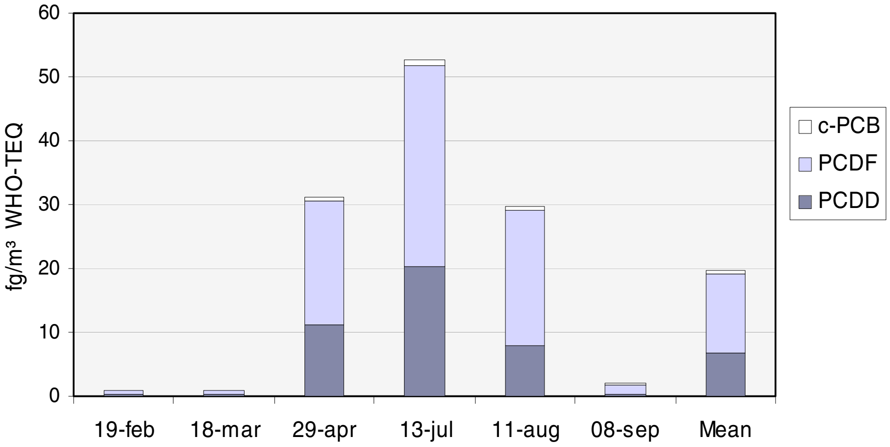

Figure 3.11 The half monthly concentrations of PCDDs, PCDFs and c-PCBs in air carried out each month in Nuuk 2004. The time indication refers to the beginning of the sample periods.

PCDD/Fs show a pronounced seasonal variation, characterised by high concentrations in the summer with a maximum in July. Otherwise, the concentrations were close to the detection limit. This annual variation profile contrasts sharply with the profiles of chlorinated pesticides in Denmark and the rest of Europe as well as in USA and Canada (Vikelsøe et al. 2005, Technical Report from NERI in press).

The steep variation with a summer maximum indicates that emission originates from local sources. Since the maximum concentrations in Nuuk occur in the summer, heating cannot be the cause and furthermore, the prevailing heating method in Nuuk is oil firing, which does not give PCDD/Fs emissions. Probably, the emissions are due to industrial activities and/or garbage burning. The local waste incinerator can be excluded as an important source since only very low concentrations of PCDD/Fs are found in flue gas and the incinerator in Nuuk is running year round.

Annual average

The total annual concentration of PCDD/Fs in Nuuk air at 19.6 fg m^{-3} WHO-TEQ is close to the annual concentration in Denmark measured at North Zealand (19.7 fg m^{-3} WHO-TEQ).

A further noteworthy feature is the very low abundance of c-PCBs in the air. There is a significant correlation between the concentrations of c-PCB and PCDD/Fs ($p = 0.02$), which indicates that they partly have the same source and partly support the above suggestion of garbage burning as the dominant source.

Biota

The low concentrations of c-PCBs in the atmosphere contrast the observations in Greenlandic marine biota (Vorkamp *et al.*, 2004) . This indicates that other sources than atmospheric long-range transport is the primary c-PCBs source in the Nuuk area. Contrary to this PCDFs dominate in the atmosphere but are insignificant in biota. These examples also highlight the need to understand the inter-compartment transport of these species as previously mentioned.

3.1.5 Nitro phenols in snow

Nitrophenols are either directly emitted from cars or produced in the atmosphere by photochemical reactions involving NOx and aromatic hydrocarbons that both are directly emitted from traffic. Nitrophenols exhibit good stability, which combined with their polarity and low vapour pressure, allows global dispersion through atmospheric transport. Long-range transport of nitrophenols to remote areas has been reported for Northern England (Luttke & Levsen, 1997). Adverse effects of nitrophenols on vegetation have been observed in remote environments, such as two alpine sites in Italy (Natangelo *et al.*, 1999). Nitrophenols have a phytotoxic effect, which occurs by inhibition of growth, transpiration and uptake of nutrients at levels of 10 µM or less.

Snow samples were collected in February 2004 and December 2004. The samples from February 2004 were not cooled during the transport to Denmark, while the samples collected in December 2004 were cooled during transport. Concentrations of nitrophenols in samples from February 2004 were a factor 10 lower than those in samples collected in December 2004. Therefore it can be concluded that nitrophenols most probably were degraded in the samples from February during the transport. Thus, concentrations of nitrophenols are only reported for the samples collected in December (Table 3.3). The relative standard deviation (RSD%) of the 3 samples gives the uncertainty of the method at the given concentration. The LOD (limit of detection) for all nitrophenols is 0.001 µg L^{-1}.

Table 3.3 Concentrations of nitrophenols (µg L^{-1}) in melted snow. The results are an average of 3 replicate samples collected in February 2005.

Compound	Average concentration µg L^{-1}	RSD%
4-nitrophenol	0.222	8.0
2,4-dinitrophenol	0.087	6.7
2,6-dinitrophenol	0.012	8.3
2,6-dimethyl- 4-nitrophenol	0.003	21.7
3-methyl-2-nitrophenol	0.029	17.2
3-methyl-4-nitrophenol	0.030	16.7

The average concentrations of nitrophenols measured in snow from Nuuk are between 5 and 30 times lower than those measured at two rural sites in Denmark (Asman *et al.*, 2005). The Danish concentrations are about 20 times higher than those measured in Antarctic snow (Vanni *et al.*, 2001). Based on laboratory studies the phytotoxicity of nitrophenols occurs at levels of 10 µM or less. The low concentrations measured in snow indicate that deposition of nitrophenols will have low or negligible effects on the Greenland vegetation, since the concentrations deposited on plants will be much below the toxicity limit.

4 Conclusion

Filter pack measurements

The values measured with filter packs were very low, and the air is very clean with respect to the pollutants in Figure 3.1 and 3.2. Receptor analysis showed that 77% of the inorganic part of the particle mass originated from sea spray, 15% from soil erosion and less than 8% from anthropogenic sources.

Mercury and ozone

GEM and ozone were close to hemispheric background values. GEM is depleted in the spring although this is observed later at Nuuk compared with Arctic sites and does not have the same strong correlation with ozone as at Arctic marine locations.

NO and NO_2

The concentrations of NO and NO_2 at the Lille Malene Station clearly show that the station is affected by local pollution. The activities around the ski lift with snow scooters and "piston bulls" are most probably the dominant source.

Chlorinated pesticides

Generally, the average annual concentrations of chlorinated pesticides in the atmosphere of Nuuk are comparable to those measured in the Eastern Canadian Arctic, indicating a degree of uniformity in contamination of the Arctic atmosphere. The concentrations of chlorinated pesticides and trifluralin are clearly influenced by air mass transport from the North American continent.

PCDD/Fs

The annual variation profile of PCDD/Fs displays a summer maximum that is suggested to come from local garbage burning. Very low levels of c-PCB concentrations were observed. They followed the annual profile of PCDD/Fs and are therefore likely to originate from the same source.

Overall conclusion

The overall conclusion is that the atmosphere in Nuuk is generally very clean apart from a small local pollution and a small contribution of long-range transport from distant anthropogenic sources located in North America.

5 Recommendations

The general low levels of pollutants (except PCDD/Fs) in Nuuk and the small contribution from sources in North America make it obvious that the monitoring activities in the Greenland atmosphere have to focus on the transport from Eurasia to Greenland. Therefore it is recommended to move the station to Station Nord located in Northeast Greenland.

Climate effects

This will also make it easier to assess the effect of climate change on the global transport of pollution, as it is in the northern Arctic that the most dramatic climatic changes are expected to take place as stated in the ACIA report (Hassol, 2004).

Model development

The general low level of pollution observed in Nuuk makes the site unfit for further validation and for improving the development of chemical-physical transport models as e.g. DEHM. This fact supports the recommendation that the Nuuk Station should be moved to a more northern position.

PCDD/Fs

The geographical distribution between west and east Greenland is at present not well described. Therefore, simultaneous measurements should be in Nuuk and at Station Nord of PCDD/Fs. This will add important information about the global atmospheric transport of these species and account for the difference observed in PCDD/Fs contamination levels in biota between east and west Greenland. Variation in climate parameters due to climate changes will also have an importance on transport and global distribution of PCDD/Fs in the Arctic.

POPs

The long-term atmospheric monitoring of POPs will further establish a relationship between POPs concentrations and the fluctuation of different climate parameters and the input of these species to the Arctic biota.

Station Nord

From a cost/benefit point of view it is preferable to move the Nuuk station to Station Nord as all infrastructure is in place; monitoring hut, electricity transport facilities. This station is located where the largest climatic effects are expected as previously mentioned and where Air masses from Eurasia initially arrive in Greenland during most winters due to a high pressure system located over the eastern part of Siberia. Finally the Danish Research Agency (SNU) is starting research activities at Station Nord, which will make it possible to coordinate monitoring activities within AMAP with research activities under SNU, which will be of mutual benefit.

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547 Contaminants in the Atmosphere

This report presents the results of atmospheric monitoring in Nuuk, Greenland. A long series of heavy metals and persistent organic Pollutants (POPs) have been measured and model calculations have been carried out supporting the interpretation of the results. Financially, the Danish Environmental Protection Agency supported this work with means from the MIKA/DANCEA funds for Environmental Support to the Arctic Region and the work is part of the Danish contribution to Arctic Monitoring and Assessment Programme, AMAP.

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