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N O R D I C W O R K I N G P A P E R S

Time series analysis of Arctic tropospheric ozone as short-lived climate force

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Preface

This report is prepared at the Danish Meteorological Institute and describes the results of the KOL project KOL-1402. The title of the project is Time Series Analysis of Arctic Tropospheric Ozone as Short Lived Climate Forcer. The project contributes to increase the knowledge in the Nordic Countries about longterm surveillance of prioritized environment indicators/climate components. An increased activity in the Arctic is to be expected in the near future with contributions from an increasing maritime traffic and an increased search for underwater occurrence of especially oil and gas. Over land is – not the least in Greenland – an increased activity to be expected by the extraction of industrial minerals. This activity will lead to an increased burning of fossil fuels and hence also an increased occurrence of short lived climate components. An increased occurrence that may only be regulated politically. In this respect especially the occurrence of VOC's and NO_x compounds are important as they represent major precursors for the formation of tropospheric ozone. The purpose of the project is to use the existing arctic time series of ozonesoundings from the nineties until today and investigate the tropospheric signal, possibly to establish a baseline for the ozone content. The implementation of the project fits well within the Nordic Environmental Management Program 2009-2012 (det nordiske miljøhåndteringsprogram 2009-2012) as it contributes to uncover the level of tropospheric ozone as a short lived climate component.

The partners in the project are the Finnish Meteorological Institute (represented by Rigel Kivi), NILU (Norwegian Institute for Air Research, represented by Georg H. Hansen) and the Danish Meteorological Institute (represented by Niels Larsen, Bo Christiansen, Ulrik Smith Korsholm and Nis Jepsen). The work has been done in 2014 and financially supported by the Nordic Council of Ministers. The stations in question are Eureka, Sodankylä, Ny Ålesund, Lerwick, Scoresbysund (Ittoqqortoormiit), Thule (Pituffik), Bear Island, Ørlandet og Gardermoen. Thanks to the Danish Environmental Protection Agency and the Danish Energy Agency for financial support of the DMI ozonesoundings in Greenland over the years. Thanks to David Tarasick (Eureka), Peter von der Gathen (Ny Ålesund) and Dave Moore (Lerwick) for the use of their ozonesounding data. Data from the other stations in question were available within the project.

Summary

An enhanced activity in the Arctic is to be expected in the near future with contributions from an increasing shipping and an increased search for underwater occurrences of especially oil and gas. Over land is – not the least in Greenland – an enhanced activity to be expected by the extraction of industrial minerals. This activity will lead to a larger burning of fossil fuels and hence also an increased occurrence of short lived climate components. An increased occurrence that may only be regulated politically.

The implementation of this project fits well into the Nordic Environment Program 2009-2012 as it contributes to reveal the level of tropospheric ozone as a short lived climate component. The project also contributes to increase the knowledge in the nordic countries on long term surveillance of prioritized environment indicators/climate components. Some of the results from the project has been incorporated in the AMAP Arctic Climate (SLCP) Issues 2015 Overview Report.

The amount of tropospheric ozone at a given location is assumed to be regulated in three ways: by local formation of precursors, by supply of precursors from other locations, and by folding of the stratosphere into the troposphere. The precursors - being they locally formed or supplied from elsewhere - may react and form tropospheric ozone. The folding of the stratosphere will directly inject ozone into the troposphere. From a policymakers point of view only the locally formed and supplied precursors may in some cases be regulated. Ozone precursors come from human activity, such as by the burning of, as well as from natural sources, such as forest fires. The stratosphere folding is a process inherent in the atmospheric dynamics. The transport of ozone precursors is the subject in a following project, Linka, which is also supported by the Nordic Council of Ministers.

Ozone sounding data series from 9 Nordic stations have been homogenized and examined. The different stations have rather different data coverage. The longest period with data is from the end of the 1980ies to 2013.

We interpolated the homogenized series to standard pressure levels and in the following analysis we focused on the tropospheric levels. A model was implied which included both a low-frequency variability in form of a polynomial, an annual cycle with harmonics, the possibility for low-frequency variability in seasonal amplitude and phasing, and noise which could be either white or AR1.

The fitting of the parameters were performed with a Bayesian approach not only giving the posterior mean values but also 95 % credible intervals.

All stations agree on a well-defined annual cycle in the free troposphere with a relatively confined maximum in the early summer.

Regarding the low-frequency variability we find that Scoresbysund, Ny Aalesund, and Sodankyla show a similar structure with a maximum near 2007 followed by a decrease. However, these results are only weakly significant. The decrease could be explained by an observed decrease in nitrogen oxide in Europe.

A significant change in the amplitude of the annual cycle was only found for Ny Aalesund. Here the peak-to-peak amplitude changed from 0.9 to 0.8 mhPa between 1995-2000 and 2007-2012.

The results were shown to be robust to the different settings of the model parameters (order of the polynomial, number of harmonics in the annual cycle, type of noise, etc). The results were also shown to be characteristic for the pressure levels in the free troposphere. Close to the surface and close to the stratosphere different processes may dominate.

Introduction

As part of the fulfillment of the Vienna Convention on protection of the ozone layer, a number of locations with long (> 20 years) time series of atmospheric profile measurements exist in the Arctic. These long time series constitutes a unique data set. The measured quantities are pressure, temperature, humidity, wind direction and wind speed as well as ozone. The measurement itself is typically being done weekly. The technical description of the ozonsonde is found below. The Vienna Convention only deals with the monitoring of stratospheric ozone. However, the time series also contains valuable measurements of tropospheric ozone as all measurements starts at the surface when the balloon is released. This tropospheric part of the soundings has not yet been analyzed. The ozone soundings constitutes the longest time series of ozone measurements in the free troposphere

Tropospheric ozone is a shortlived climate forcer (SLCF) or shortlived climate component (SLCC). The purpose of the project is to use the long time series from Sodankylä in Finland, Ny Ålesund in Spitsbergen, Scoresbysund and Thule in Greenland, and the somewhat shorter time series from Bear Island, Ørlandet and Gardermoen in Norway, to investigate the tropospheric content of ozone to establish a baseline for the arctic tropospheric ozone content during the recent 25 years. Additional data from a Canadian station is used in order to cover as much of the Arctic as possible.

Due to the diminishing amount of sea ice in the Arctic the activity – none the less at sea – has increased in recent years. This encompasses an increased burning of fossile fuels and hence also an increased release of NO_x-gasses and VOX which inevitably will lead to an increased content of tropospheric ozone. Establishing a baseline for tropospheric ozone is considered important to determine a useful and realistic arctic monitoring project for shortlived climate components.

In Figure 1 [Guicherit et al., 2000] is shown an example of an annual tropospheric ozone budget. The contributions to the balance are: ozone production based on precursor emission, ozone destruction, influx from the stratosphere, transport and either wet or dry deposition.

Figure 1 - Tropospheric ozone sources

The anthropogenic pollution from the Northern Hemisphere is the dominant source of O₃ and HNO₃ in the Arctic at pressures greater than 400 hPa, and the stratospheric influence is the principal contribution at pressures less 400 hPa [Wespes et al., 2012]. The anthropogenic sources may either be formed in situ or transported to the site of reaction. Nitrogen oxides are considered especially important in this respect. Besides from being formed by anthropogenic activities nitrogen oxides may also be formed in lightning processes [Cairo et al., 2010]. The chemical ozone production takes place in the troposphere only while the chemical ozone destruction mechanism may be the same as is seen in the stratospheric vortex [Larsen et al., 1994]. Aerosols act as catalysts upon which the breakdown takes place. In the stratosphere these aerosols may be nitric acid or ice particles [Larsen et al., 1996]. The dry deposition rate has been calculated to be 0.1-0.5 cm/s [Pio et al., 1996] depending on time of year and time of day. These values were estimated for Southern Europe. Here, the concept of the deposition being composed of additive contributions of aerodynamic resistance, quasi-laminar boundary layer resistance and canopy resistance, is introduced. However, similar values have been obtained for Western Alaska, that is 0.12-0.24 cm/s [Jacob et al., 2012]. The average lifetime for tropospheric ozone is 19-33 days [IPPC, 2007].

The influx from the stratosphere may be caused by tropopause folding. This has been demonstrated using backwards trajectory calculation [Sørensen et al., 2000]. Synoptic scale processes as represented by the 250 hPa geopotential height have also been successfully linked to the recent ozone increases in the lowermost stratosphere [Harris et al,

2008]. Another influx can be simple transport of low ozone air parcels. This has been demonstrated using NWP [Rasmussen et al., 1997].

1. Included stations

The included stations are listed in the following Table 1. These stations have all been submitting data to either the NILU and/or the NDACC database over the years. The geographic distribution is shown in Figure 2. The number of soundings for each station as a function of year is shown in Figure 3.

Station Name	Latitude	Longitude
Eureka (CA)	80.1°N	86.4°W
Ny Aalesund (DE)	78.9°N	11.9°E
Thule [Pituffik] (DK)	76.5°N	68.7°E
Bear Island (N)	74.3°N	19.0°E
Scoresbysund [Ittoqqortoormiit] (DK)	70.5°N	22.0°E
Sodankylä (FI)	67.4°N	26.7°W
Orland (N)	63.7°N	9.6°E
Gardermoen (N)	60.2°N	11.1°E
Lerwick (UK)	60.1°N	1.2°W

Table 1 – Included stations sorted by Latitude

Figure 2 – Geographic coverage of included stations

Figure 3 – Number of Soundings per year per Station

For those stations participating in the yearly Match Campaign run by AWI [Rex, 1993; von der Gathen et al., 1995], more ozonesondes are released in late winter-early spring. Thule are recently only launching ozonesondes during the Match Campaign, hence the low number of sondes per year.

2. The ozonesonde

The ozonesonde is an electrochemical device containing two electrode chambers: an anode chamber filled with potassium iodide saturated phosphate buffer and a cathode chamber filled with same phosphate buffer containing a well defined concentration of potassium iodide. During ascent through the atmosphere a constant volume pump is drawing atmospheric air through the cathode chamber. The content of ozone in the air sample is reacting with the potassium iodide and gives rise to a current proportional to the ozone amount. The electrode chambers and the pump is installed in a Styrofoam box for insulation purposes. To keep the buffer liquids from freezing during ascent a simple heater element, that is batteries being discharged through a resistor, is keeping the temperature in box at 10-25°C. A thermistor is sensing the actual temperature inside the box. On the outside of the Styrofoam box a regular radiosonde is mounted. The radiosonde is measuring pressure, temperature, humidity and wind speed and wind direction during ascent. The ozone current and the box temperature is via an interface transmitted to a ground receiver along with the radiosonde parameters. The styrofoam box containing the ozone sonde and the radiosonde is lifted with a helium filled rubber balloon. In good conditions the balloon may reach an altitude at 35-40 km. Typical vertical resolution is 30 m.

A sample launch is shown in Figure 4.

Figure 4 – Launch of ozonesonde from Scoresbysund on Dec. 5th 2013

In the figure the green line is the humidity, the black line is the temperature inside the ozonesonde, the red line is the air temperature and the blue line is the ozone value. As is seen, around the tropopause the air dries out, the temperature begins to rise as does the ozone value.

3. Methodology

3.1 Homogenization of data series

Different types of ozonesondes have been in use over the years, the primary two types being ENSCI and Science Pump. Both types are constructed as described in Chapter 3. And for each ozonesonde type there is a recommended composition of the anode and cathode buffers in use. As well as for the concentration of potassium iodide. Problems arise if one wants to change to a different brand of ozonesonde. Changing the sonde as well as the buffer may lead to confusion amongst the staff doing the actual launches. Therefore many launches have been made using buffers recommended for Science Pump ozonesondes in ENSCI type ozonesondes. To investigate the difference between the two sonde types and buffer types a number of in vivo measurements were used, for instance [Kivi et al., 2008] and [BESOS, 2004], along with experimental setup measurements [JOSIE, 2000]. The outcome from the comparison runs provided recommendations and transfer functions for the conversion from one ozonesonde type using a specific buffer composition to the one giving the agreed upon result [Smit et al., 2012]. Software was made to run through the Nasa Ames 2160 [Gaines et al., 1998] formatted data files to identify the type of ozonesonde and buffer type used, and recalculate the data if necessary according to the required transfer function.

3.2 Model description

For each station and for each homogenized ozone sounding the ozone is interpolated to standard pressure levels 900, 800, ... 300, 250, ... 100, 80, 70 ... 10 hPa. The resulting ozone fields are shown as function of time and pressure in Figure 5 for each station. Here and in the rest of the report we measure ozone with its partial pressure and use the unit of mPa. We are mainly here interested in the troposphere and time-series of ozone at 500 hPa are shown in Figure 6. We note that in general the data are highly irregular with large gaps but also that the details vary a lot among the stations. We also note that the ozone records have large annual cycles and also a considerable amount of scatter.

At each pressure level we want to model the temporal development of ozone. We are particularly interested in potential low-frequency trends and changes. We therefore use a model that contains a trend, an annual cycle and noise. The model has the form

$$y = \lambda_0 + \lambda_1 t + \lambda_2 t^2 + \dots a_1 \sin(2\pi t + \theta_1) + a_2 \sin(2\pi 2t + \theta_2) \dots + \xi \quad (1)$$

We note the following properties:

- The trend consists of a constant λ_0 , a linear trend $\lambda_1 t$, and higher order polynomials up to $\lambda_{n_{pol}-1} t^{n_{pol}-1}$.
- The annual cycle consist of a sum of n_{cycle} sinusoidals $a_i \sin(2\pi i t + \theta_i)$ with frequency 1, 2, 3 ... n_{cycle} . The higher harmonics allow the seasonal cycle to be asymmetric. The amplitudes and phases of the cycles have trends with n_{trend}^a and n_{trend}^θ terms: $a_i = a_{i,0} + a_{i,1}t + \dots a_{i,n_{trend}^a} t^{n_{trend}^a}$, $\theta_i = \theta_{i,0} + \theta_{i,1}t + \dots \theta_{i,n_{trend}^\theta} t^{n_{trend}^\theta}$. This allows the annual cycle to change over time.
- The noise is either independent Gaussian with variance σ^2 or an auto-regressive order 1 process with AR1 coefficient θ and variance σ^2 .
- Then, the model totally includes $2 + n_{pol} + 2n_{cycles}(1 + n_{trend})$ parameters under AR1 noise and one less under Gaussian noise.

Figure 5 - Ozone partial pressure [mPa] as function of time and pressure for the 9 stations

The model is nonlinear and includes a considerable number of parameters. We therefore choose a Bayesian approach for inference. Samples from the posterior are obtained by a simple Metropolis-Hastings algorithm and we assume flat priors for all parameters. This approach produces ensembles of all parameters, all parts of the model, and all derived quantities. These ensembles give the posterior distributions of the quantities under consideration. From these distributions we calculate and report the posterior mean and the 95 % credible (confidence) intervals. We also note that the Bayesian approach does not require regular temporally gridded data which is fortunate given the highly irregular distributed ozone data.

Figure 6 - Ozone at 500 hPa (partial pressure mPa) for the 9 stations

4. Results

4.1 Trends and annual cycle

Given the large differences in data coverage among the different stations we can not expect that all station can provide sufficient information to constrain models with a high number of parameters. We therefore begin the analysis with a simple version of the model including only the polynomial trend and a fixed annual cycle. Figure 7 shows for 500 hPa the raw data (black points), posterior mean of the the non-stochastic part of the model, i.e., the polynomial part and the annual cycle (cyan), and posterior mean of the polynomial part of the model (green) alone.

Figure 7 - Observations (black), model mean fit (cyan) and polynomial part of the model (green) as function of time at 500 hPa. Model setting: $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^a = n_{trend}^\theta = 0$, and white noise

It is obvious that the Bayesian procedure has produced reasonable fits dominated by an annual cycle and including a weak inter-decadal variability. It is also obvious that there is a considerable residual scatter at all stations. This scatter is the expression of dynamical and chemical processes in the atmosphere as well as measurement noise.

Figure 8 - The annual cycle as function of day of year at 500 hPa. Black curve shows posterior mean, colored curves indicate the 95 % credible intervals for each day of year. Model setting: $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^1 = n_{trend}^0 = 0$ and white noise

As mentioned the Bayesian approach not only give a point values but the whole posterior distributions. Figure 8 shows both the mean annual cycle as well as the 95 % credible interval for each day of year. The annual cycle has a surprising similarity for all stations. It has a minimum in winter, a maximum in early summer and a peak-to-peak amplitude of approximately 1 mPa. We also note that the annual cycle would not be well modelled with a single sinusoidal as the early summer peak is more temporal confined than the winter minimum. The width of 95 % credible

intervals reflects the data coverage and are largest for Thule, Gardermoen and Orland.

Figure 9 - The polynomial part of the model as function of time at 500 hPa. Green curve shows posterior mean, black curves indicate the 95 % credible intervals for each point in time. Model settings: $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^a = n_{trend}^0 = 0$ and white noise

Figure 9 shows both the mean polynomial part of the model (the cyan curve in Fig. 3) and the 95 % credible interval for each point in time. For all stations the long term background value is around 3 mPa and the polynomial part is relatively flat with some weak low-frequency variability. The 95 % credible intervals are quite large relative to the low-frequency variability. It again mainly reflects the data coverage but the credible intervals also increases near the beginning and end of the time-series where data are limited because of the asymmetry. For Scoresbysund, Sodankyla, Ny Aalesund, and Eureka some significant albeit weak low-frequency variability can be discerned. At Scoresbysund the ozone partial pressure increases until a maximum is reached near

2007 followed by a weak decrease. Ny Aalesund shows similar behavior but now with the maximum around 2003. Sodankyla also shows a decrease in recent years with a maximum around 2005. However, Eureka show a qualitative different variability with a strong increase from 1990 to 2000 followed by a quiet period until 2008 after which an increase is found again.

Figure 10 - Time varying annual cycle at 500 hPa. Model settings: $n_{pol} = 4$, $n_{cycles} = 3$, $n_{trend}^a = 2$, $n_{trend}^\theta = 0$, and white noise Changes in the annual cycle

We saw in the last section that the annual cycle was well modelled and almost identical for all stations. This provides some hope for that we have enough information to detect potential changes in the annual cycle. We limit the following analysis to the four stations with best data coverage: Scoresbysund, Sodankyla, Ny Aalesund, and Eureka.

To study the temporal variability in the annual cycle we now apply a model with $n_{trend}^a = 2$ i.e. we allow the amplitudes of the components of the annual cycle to vary in time like a second order polynomial. In this model we keep the phases of the components of the annual cycle fixed.

Figure 10 shows the now time-varying annual cycles. For Sodankyla the annual cycle seems pretty constant while for Scoresbysund and Eureka the annual cycle is largest before 1995. The most pronounced variability is found at Ny Aalesund where the annual cycle peaks near year 2000 and decreases thereafter.

It should be understood that there are large uncertainties connected to the changes in the annual cycle. This is seen in Figure 11 where the annual cycles averaged over 1995-2000 and 2007-2012 are shown together with their 95 % credible intervals for each day of the year. It is clear that the only significant change is found at Ny Aalesund which shows a slight, significant decrease from 0.9 to 0.8 in the peak-to-peak amplitude.

In the models fit above the phase of the annual components were kept fixed. Allowing the phases to vary in time like a second order polynomial, as for the amplitude, makes it possible to investigate if there is a drift of the seasonality in the ozone. It also help to establish the robustness of the results in Figure 11. In Figure 12 the results similar to those

in Figure 11 are shown, but now with $n_{trend}^{\theta} = 2$ instead of $n_{trend}^{\theta} = 0$. We see that there is very little and insignificant change in the phase of the annual cycle and that the results about the amplitudes are very robust.

Figure 11 - Average annual cycles over 1995-2000 (cyan) and 2007-2012 (red) at 500 hPa. Full curve is the posterior mean, dashed curves indicate the 95 % credible intervals. Model settings: $n_{pot} = 4$, $n_{cycles} = 3$, $n_{trend}^a = 2$, $n_{trend}^{\theta} = 0$ and white noise

4.2 Vertical variations

The discussions in the previous sections have focused on the 500 hPa layer. In this section we briefly discuss the results of modelling the ozone at other levels. We will focus on Scoresbysund for the low-frequency variability and Ny Aalesund for the annual changes as these were the cases where we found the most interesting results in the previous sections.

Figure 13 show the posterior mean model fit at Scoresbysund for four pressure levels: 900, 700, 500 and 300 hPa. At all four pressure levels the low-frequency variability is similar with the maximum near 2005. The largest difference is seen at 300 hPa where there is a very strong scatter and an obviously positively skewed distribution. This behavior is due to the proximity of the 300 hPa layer to the stratosphere. The positive excursions are related either to variation of the tropopause height or to intrusions of ozone rich stratospheric air into the troposphere.

The mean annual cycle at Ny Aalesund is shown in Figure 14 for the four pressure levels. The seasonal cycles at 500 and 700 hPa are almost similar. At the near-surface (900 hPa) the annual cycle is qualitatively different with an extra maxima in late autumn. Also, the primary maximum appears a couple of months earlier than in the middle troposphere. At 300 hPa the amplitude of the annual cycle is approximately a factor of two larger than at the other levels; this is probably again an effect of the proximity to the stratosphere.

Figure 12 - Average annual cycles over 1995-2000 (cyan) and 2007-2012 (red) at 500 hPa. Full curve is the posterior mean, dashed curves indicate the 95 % credible intervals. Model settings: $n_{pol} = 4$, $n_{cycles} = 3$, $n_{trend}^a = n_{trend}^0 = 2$, and white noise

The difference in the average annual cycle over 1995-2000 and 2007-2012 at Ny Alesund is seen in Figure 15. The decrease in annual cycle is seen at 700 hPa as well as at 500 hPa. However, the situation is different at 900 hPa where there might even be a small, weakly significant increase. At 300 hPa the 95 % credible intervals are too large for any change to be seen.

Figure 13 - Observations (black), model mean fit (cyan) and polynomial part of the model (green) as function of time at Scoresbysund. Panel a) 900 hPa, b) 700 hPa, c) 500 hPa, d) 300 hPa. Model settings: $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^a = n_{trend}^0 = 0$, and white noise

Figure 14 - The annual cycle as function of day of year at Ny Aalesund. Black curve shows posterior mean, colored curves indicate the 95 % credible intervals for each day of year. Panel a) 900 hPa, b) 700 hPa, c) 500 hPa, d) 300 hPa. Model settings: $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^a = n_{trend}^g = 0$, and white noise

4.3 Robustness of the results

Our model allows for many different settings of the parameters and it is not obvious which is the optimal choice. We have, for example, in the previous discussion restricted ourselves to model setups with white noise.

In this section we briefly discuss the robustness of the results to changes in the parameters. We will restrict the presentation to Scoresbysund for the low-frequency variability and to Ny Aalesund for the changing annual cycle but similar results are found at other stations.

Figure 16 shows the polynomial part of the model for Scoresbysund at 500 hPa for seven different model settings, including settings with white noise or AR1 noise, and including settings with different orders of the polynomial trend. We observe that all model settings agree on the shape of the low-frequency variability, in particular they agree on the maximum obtained around year 2005.

Figure 15 - Average annual cycles over 1995-2000 (cyan) and 2007-2012 (red) at Ny Aalesund. Full curve is the posterior mean, dashed curves indicate the 95

% credible intervals. Panel a) 900 hPa, b) 700 hPa, c) 500 hPa, d) 300 hPa. Model settings: $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^a = 2$, $n_{trend}^\theta = 0$, and white noise

Figure 17 shows the average cycles over 1995-2000 and 2007-2012 for three different model settings including settings with white noise or AR1 noise, and including settings with different numbers of seasonal harmonics. Again we observe that all model settings agree on the slight change in the annual cycle.

These results are typical for the stations with best data coverage. Some sensitivity is seen for stations with large gaps between soundings. In some cases with few data and many parameters, in particular when we choose AR1 noise, the Monte Carlo approach does not converge.

Figure 16 - The polynomial part of the model as function of time for different parameter settings. Green curve shows posterior mean, black curves indicate the 95 % credible intervals for each point in time. Scoresbysund, 500 hPa. From top: $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^a = n_{trend}^\theta = 0$, and white noise, $n_{pol} = 4$, $n_{cycles} = 2$, $n_{trend}^a = n_{trend}^\theta = 0$, and AR1 noise, $n_{pol} = 4$, $n_{cycles} = 3$, $n_{trend}^a = 2$, $n_{trend}^\theta = 0$, and white noise, $n_{pol} = 4$, $n_{cycles} = 3$, $n_{trend}^a = n_{trend}^\theta = 2$, and white noise, $n_{pol} = 4$, $n_{cycles} = 1$, $n_{trend}^a = n_{trend}^\theta = 1$, and AR1 noise, $n_{pol} = 4$, $n_{cycles} = 3$, $n_{trend}^a = n_{trend}^\theta = 2$, and AR1 noise, $n_{pol} = 6$, $n_{cycles} = 2$, $n_{trend}^a = n_{trend}^\theta = 1$, and AR1 noise

Figure 17 - Average annual cycles over 1995-2000 (cyan) and 2007-2012 (red) for Ny Aalesund at 500 hPa. Full curve is the posterior mean, dashed curves indicate the 95 % credible intervals. From top: $n_{pol} = 4$, $n_{cycles} = 3$, $n_{trend}^a = n_{trend}^\theta = 2$, and white noise, $n_{pol} = 4$, $n_{cycles} = 1$, $n_{trend}^a = n_{trend}^\theta = 1$, and AR1 noise, $n_{pol} = 4$, $n_{cycles} = 3$, $n_{trend}^a = n_{trend}^\theta = 2$, and AR1 noise

5. Conclusions and discussion

We have collected, homogenized and analyzed ozone soundings from 9 Nordic stations. Our main findings are:

- The different stations have very different data coverage. The longest period with data is from the end of the 1980ies to 2013.
- We interpolated the homogenized series to standard pressure levels and in the following analysis we focused on the tropospheric levels.
- We applied a model which included both a low-frequency variability in form of a polynomial, an annual cycle with harmonics, the possibility for low-frequency variability in seasonal amplitude and phasing, and noise which could be either white or AR1.
- The fitting of the parameters were performed with a Bayesian approach not only giving the posterior mean values but also 95 % credible intervals.
- All stations agree on a well-defined annual cycle in the free troposphere with a relatively confined maximum in the early summer.
- Regarding the low-frequency variability we find that Scoresbysund, Ny Aalesund, and Sodankyla show a similar structure with a maximum near 2005 followed by a decrease. However, these results are only weakly significant.
- A significant change in the amplitude of the annual cycle was only found for Ny Aalesund. Here the peak-to-peak amplitude changed from 0.9 to 0.8 mhPa between 1995-2000 and 2007-2012.
- The results were shown to be robust to the different settings of the model parameters (order of the polynomial, number of harmonics in the annual cycle, type of noise, etc).
- The results were also shown to be characteristic for the pressure levels in the free troposphere. Close to the surface and close to the stratosphere different processes may dominate.

Earlier work covering data from 1989-2003 [Kivi et al., 2007] suggests a linear increase in the free troposphere of $11.3 \pm 1.8\%$ over the 15 years of observation, consistent with our observations for Thule, Scoresbysund, Ny Ålesund, Eureka and Orland. Looking at the January to April period an even more pronounced increase of $16.0 \pm 3.1\%$ over the 15 years is observed. The observed change is suggested to be due to changes in the Arctic Oscillation.

In another study covering longer-term, ie 20-40 years, observations, no overall change in tropospheric ozone is found for the included stations north of 60°N (Resolute) [Oltmans et al., 2004]. The data are aver-

aged into layers: surface-850 hPa, 850-700 hPa, 700-500 hPa and 500-300 hPa. Cubic polynomial fit is used for the regression.

For the Canadian stations covering some 20 years of data from 1981-2001 an negative linear trend is found for the tropospheric as well as for the lower stratospheric ozone levels [Tarasick et al., 2005].

The polynomial part of the Bayesian fit shows a significant decrease in tropospheric ozone from around 2007 for the stations Orland, Thule, Scoresbysund and Sodankylä. For Thule the decrease is less pronounced, and the data is more sparse at this station. This decrease may be explained by the corresponding decrease in nitrogen oxide level as is seen in the Euro Zone. Current levels are 50 % of 1990 level [EEA, 2014]. And nitrogen oxide is an important precursor for the production of tropospheric ozone, suggesting a long transport of this species.

For the stations Lerwick and Ny Ålesund the levels seems to be stable throughout the observation period. For Eureka a slight increase has been observed since 2007.

The timeseries for Gardermoen and Bear Island are rather short but interestingly points in different directions despite their short geographic distance (around 500 km).

In future work we will try to relate the changes to dynamical variability in the atmosphere such as changes in the NAO, the polar front, or transport of chemically active species.

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Sammenfatning

Der forventes fortsat øget aktivitet i Arktis i de kommende år. Både fra en voksende skibstrafik og fra en øget eftersøgning efter undersøiske forekomster af især olie og gas. Til lands forventes der – ikke mindst i Grønland - ligeledes øget aktivitet ved udvinding af industrimineraler. Denne aktivitet vil medføre en større afbrænding af fossile brændsler og dermed også en øget forekomst af kortlivede klimakomponenter. En øget forekomst, der kun kan reguleres ad politisk vej.

Ozonsonderingsdata fra 9 nordiske stationer er blevet homogeniseret og undersøgt. De forskellige stationer har meget forskellig datadækning. Den længste periode med data er fra slut 1980'erne til 2013.

Vi har interpoleret de homogeniserede dataserier til standardtrykniveauer, og i den følgende analyse har vi fokuseret på de troposfæriske niveauer. Der blev anvendt en model, som indbefattede både en lavfrekvent variabilitet i form af et polynomium, en årlig cyklus med harmoniske led, mulighed for lavfrekvent variabilitet i sæsonmæssig amplitude og fase, og støj der kan være enten hvid eller AR1 (autoregressiv af første orden).

Tilpasning af parametrene blev udført ved en bayesiansk fremgangsmåde, der ikke alene giver posterior middelværdier men også 95 % bayesianske confidensintervaller.

Alle stationer udviser en veldefineret årlig cyklus i den frie troposfære med et relativt begrænset maksimum i den tidlige sommer.

Med hensyn til den lavfrekvente variabilitet har vi fundet, at Scoresbysund, Ny Aalesund og Sodankylä udviser en ensartet struktur med et maksimum omkring 2007 efterfulgt af et fald. Disse resultater er imidlertid kun svagt signifikante. Faldet kan forklares ved et observeret fald i nitrogenoxid over Europa.

En signifikant ændring i amplitude af den årlige cyklus blev kun observeret for Ny Aalesund. Her ændrede amplitude sig fra 0,9 til 0,8 mhPa mellem 1995-2000 og 2007-2012.

Resultaterne viste sig at være robuste i forhold til de forskellige indstillinger af modelparametrene (polynomieorden, antal harmoniske i den årlige cyklus, støjtype osv.). Resultaterne viste sig også at være karakteristiske for trykniveauerne i den frie troposfære. Tæt på overfladen og tæt på stratosfæren kan forskellige processer dominere.