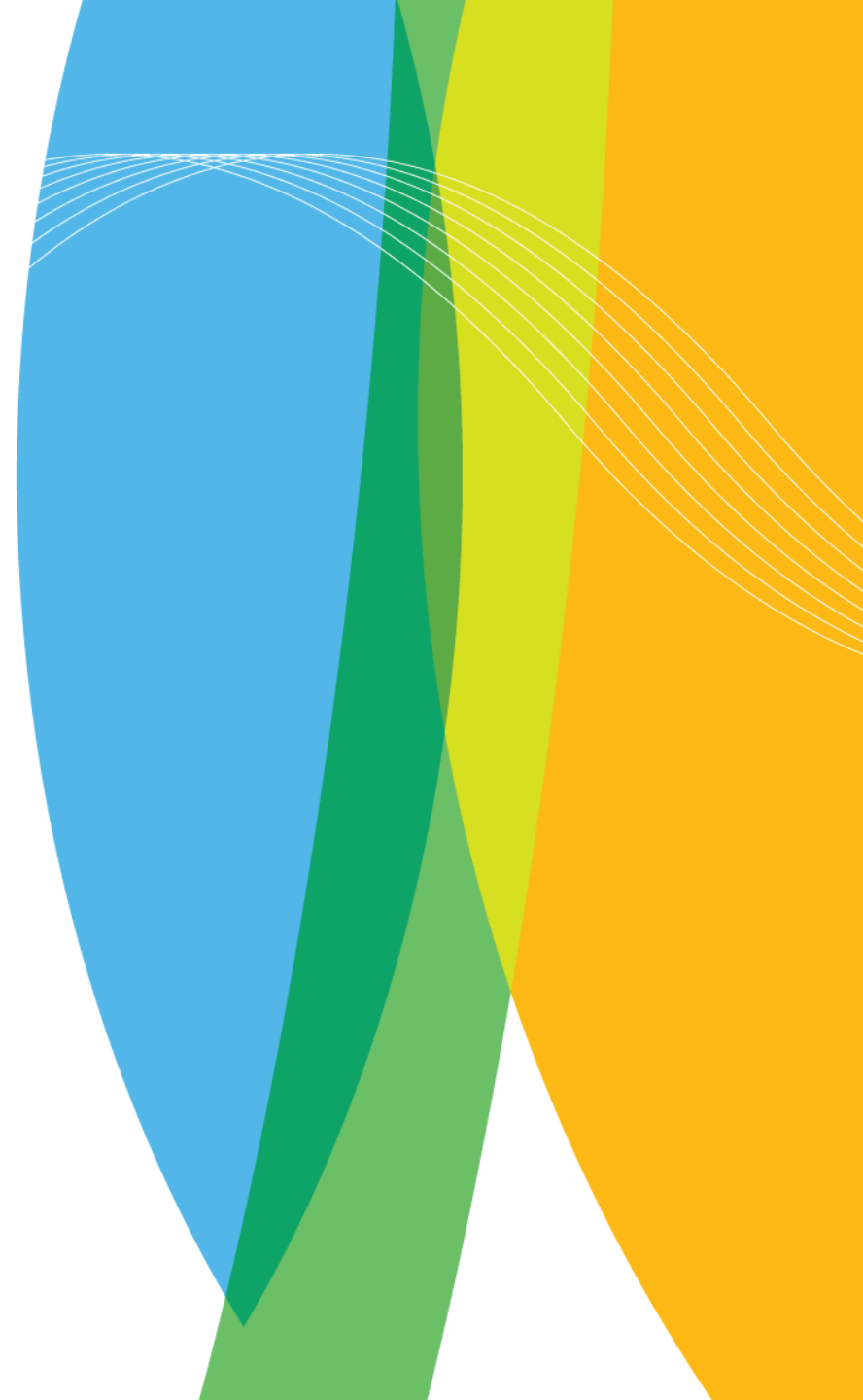




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A method for detection of local agricultural effects in time series of ammonia+ammonium at EMEP stations

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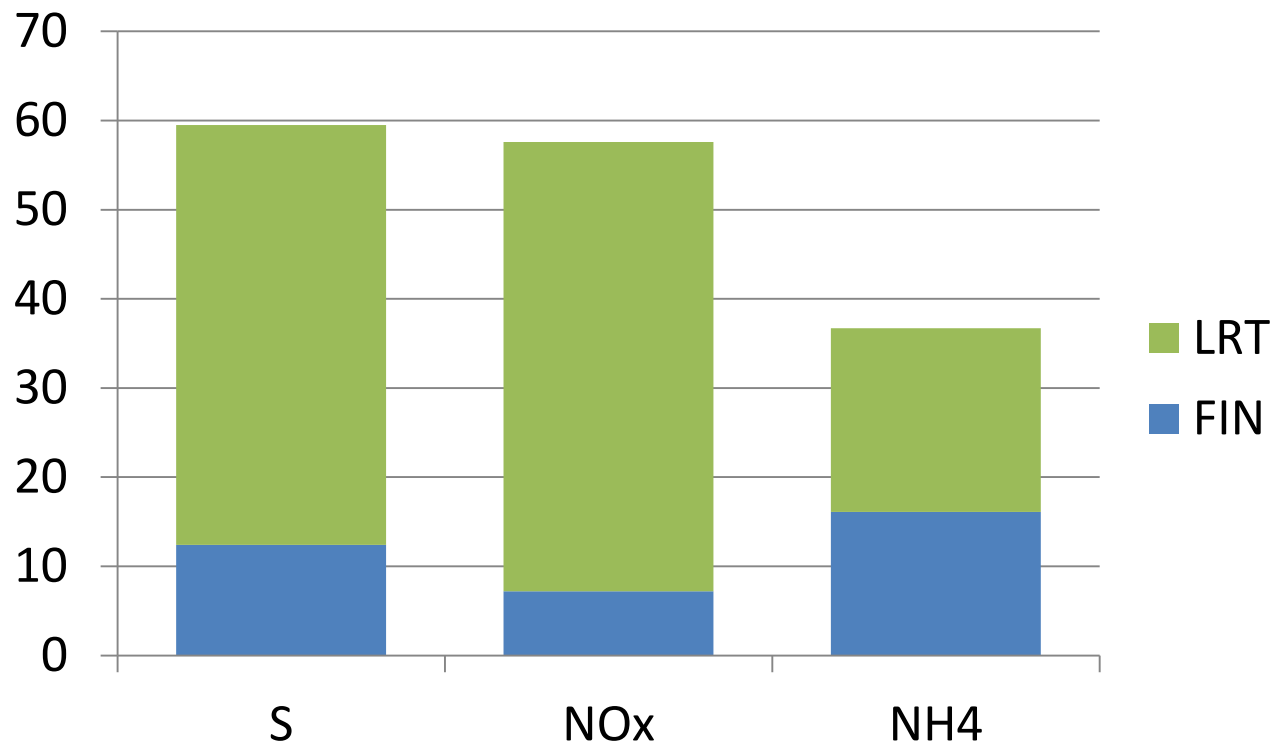


Introduction

- main contributor to Finnish air quality in background areas is LTR
- local emission sources might occasionally contribute substantially
- need to examine local and regional differences



The atmospheric load of S and N in Finland is mostly long range transported



Origin of deposition in 2008, kt S or N. EMEP 2010.

Material

- seven-year time series of atmospheric monitoring data for SO_2 , SO_4^{2-} , $\text{NO}_3^- + \text{HNO}_3$, $\text{NH}_4^+ + \text{NH}_3$ and Cl^-
- same methods
- from stations at Ähtäri (left) and Hyytiälä (right) in southern Finland
- located 85 km apart



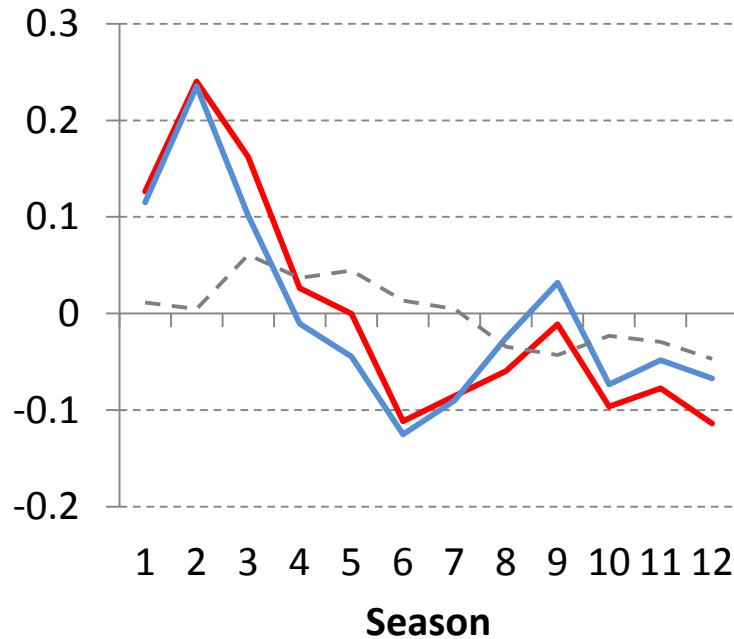
Methodology

- Sampling (filter pack) and analysis (ion chromatography) according to UN/ECE EMEP Guidelines
- Daily (2003) or weekly (2004-2009) sampling at Ähtäri, 3 samples (2+2+3 days)/week at Hyytiälä
- Expanded measurement uncertainty $U_{\text{tot}} < 5\%$ (Karlsson et al. 2007)
- Good agreement between daily and weekly sampling
- Land use around stations mostly forest, also some arable land, wetlands, a lake

Statistical analysis

- Generalized least-squares (GLS) regression with classical decomposition and autoregressive moving average (ARMA) errors
- Method of Brockwell and Davies (2002) (ITSM Professional 7.3, B and D Enterprises Inc., 2005), applied by Anttila and Tuovinen (2009) and Ruoho-Airola (2012)
- estimation of the seasonal components by moving average method
- subtraction of the seasonal components from the time series
- calculation of the linear trend by GLS-ARMA –method
- estimation of the optimal ARMA-model to the residuals by iteration

Comparison of monthly mean values, example SO₄

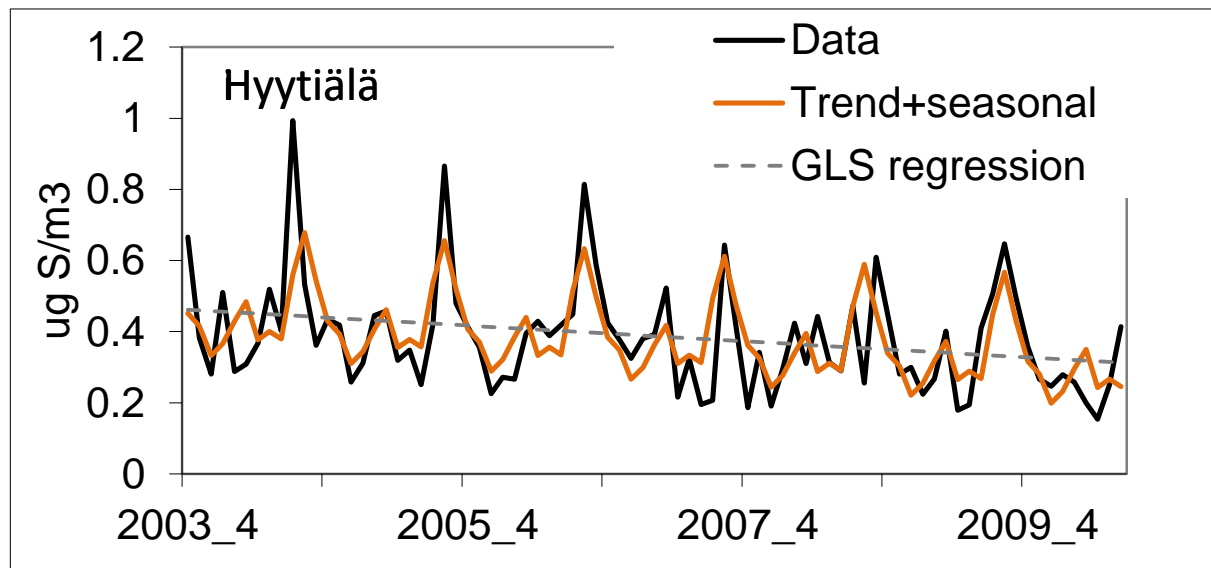
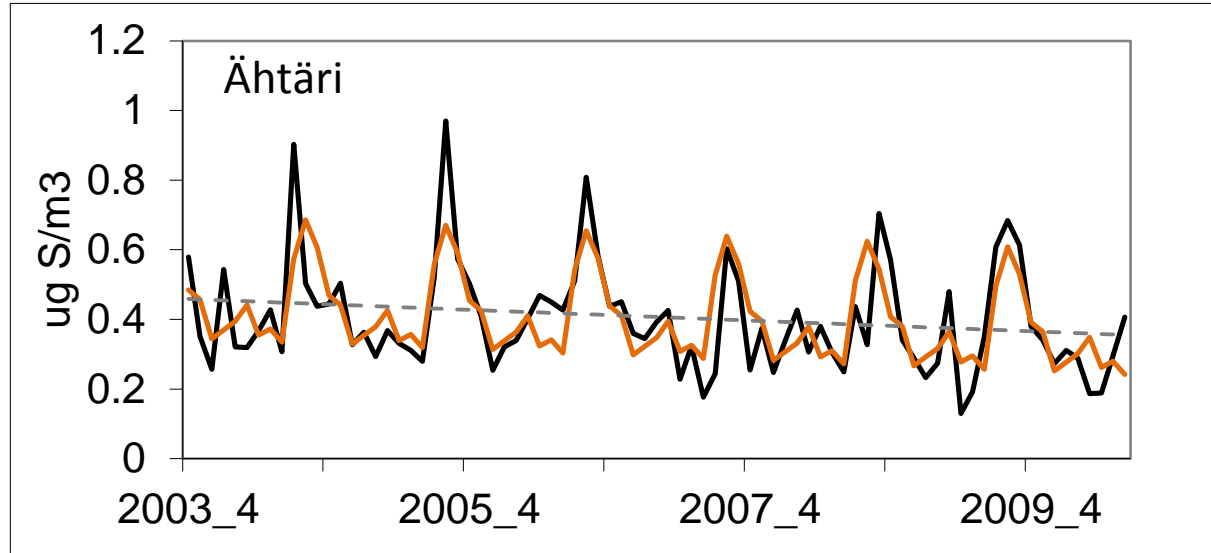


Only small difference in seasonal components, between the stations.

— Ähtäri — Hyytiälä - - - Diff.

Station	Order of ARMA model (p,q)	Trend, %/year
Ähtäri	4,2	-3.4 ± 2.4
Hyytiälä	1,1	-4.8 ± 1.2

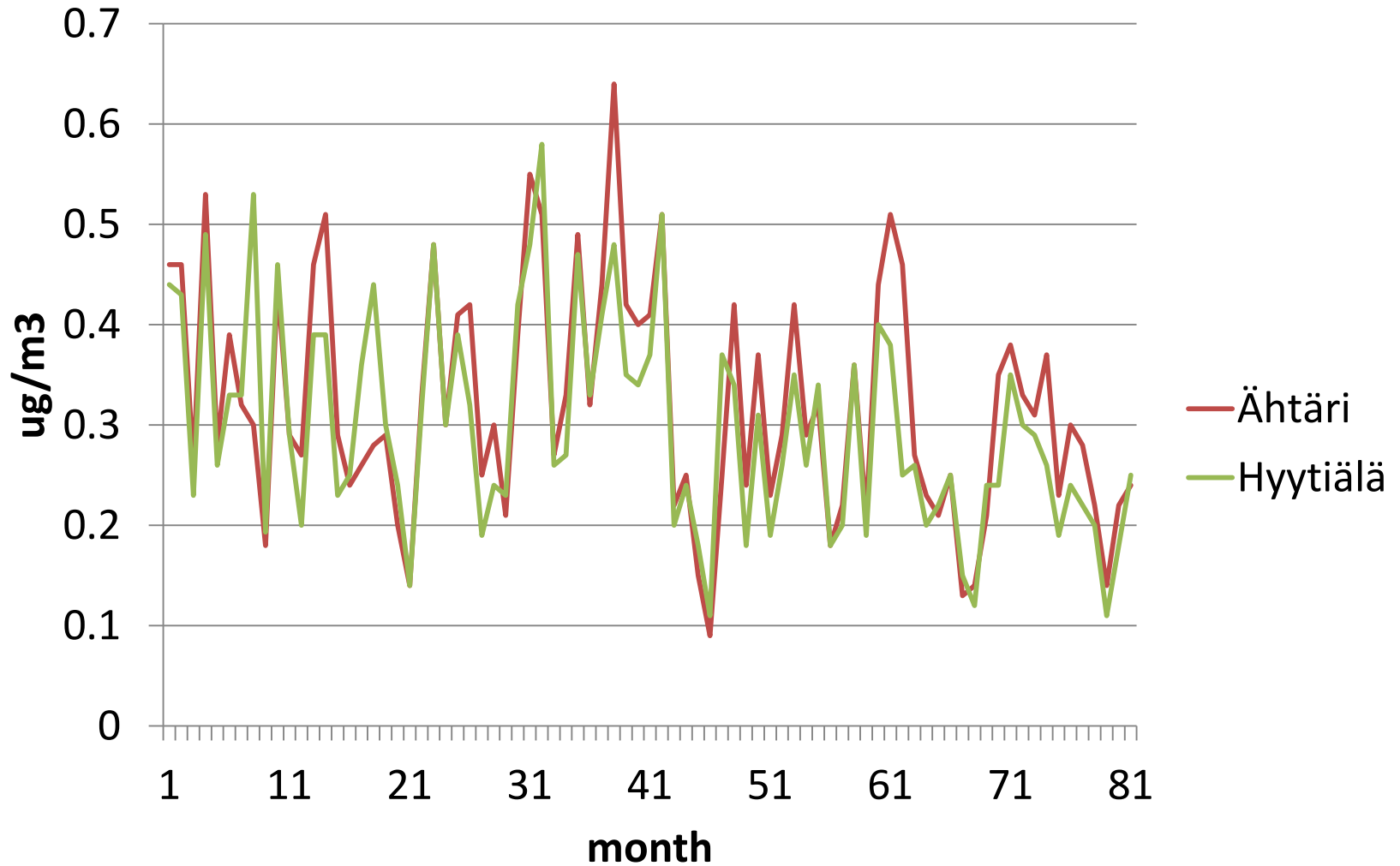
The seasonal component + trend explained well of the time series



The SO_4 time series at Ähtäri and Hyytiälä can be explained by LTR, no remarkable local effect available

Instead, the NH_3+NH_4 time series differ at the two stations

Atmospheric ($\text{NH}_3 + \text{NH}_4$) concentration, monthly mean values



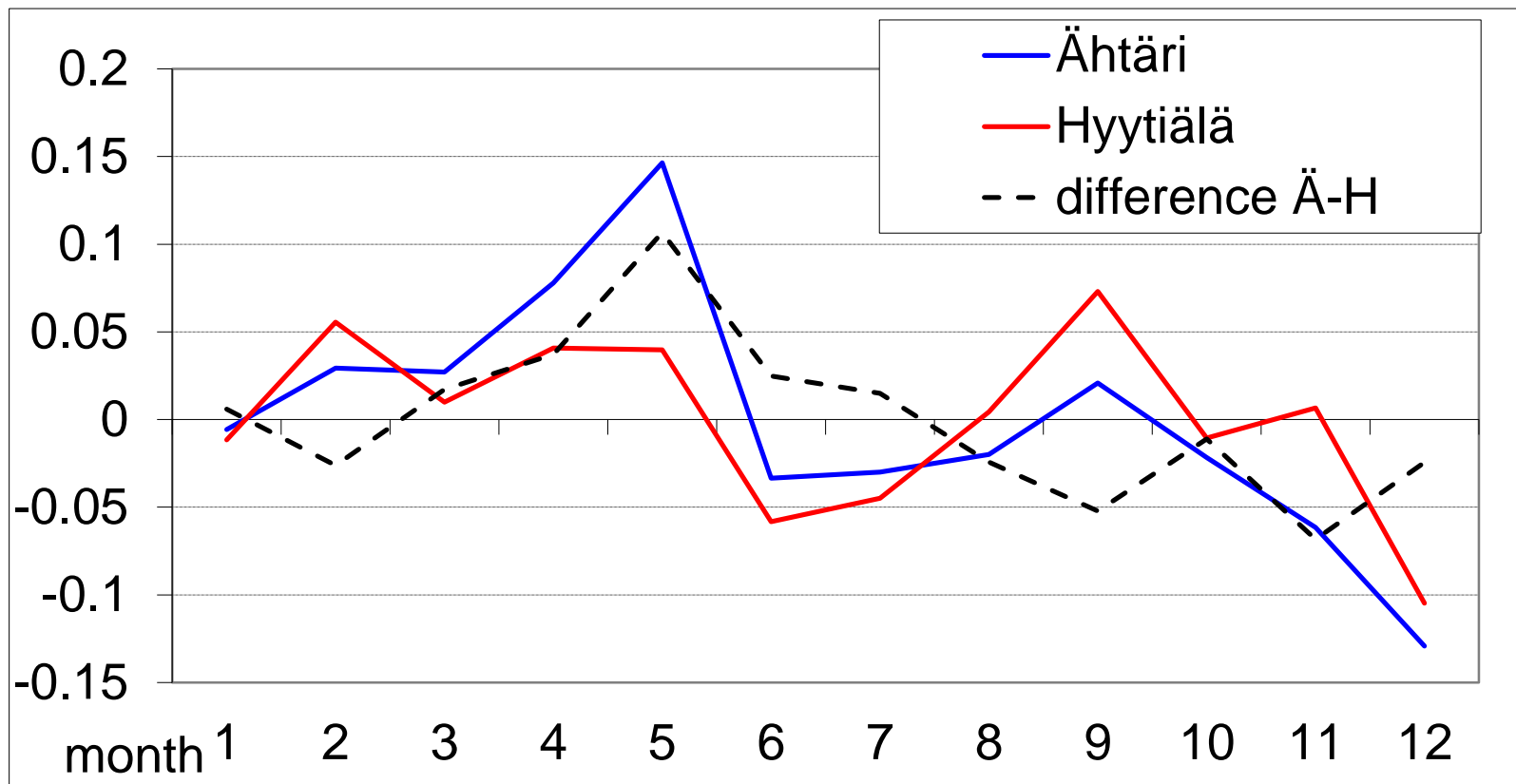
The trends in the deseasonalized time series of NH_3+NH_4 in Hyytiälä and Ähtäri overlap

Ähtäri $-3.6 \pm 3.4 \%$

Hyytiälä $-5.8 \pm 2.6 \%$

so, the seasonal components might include the local influence

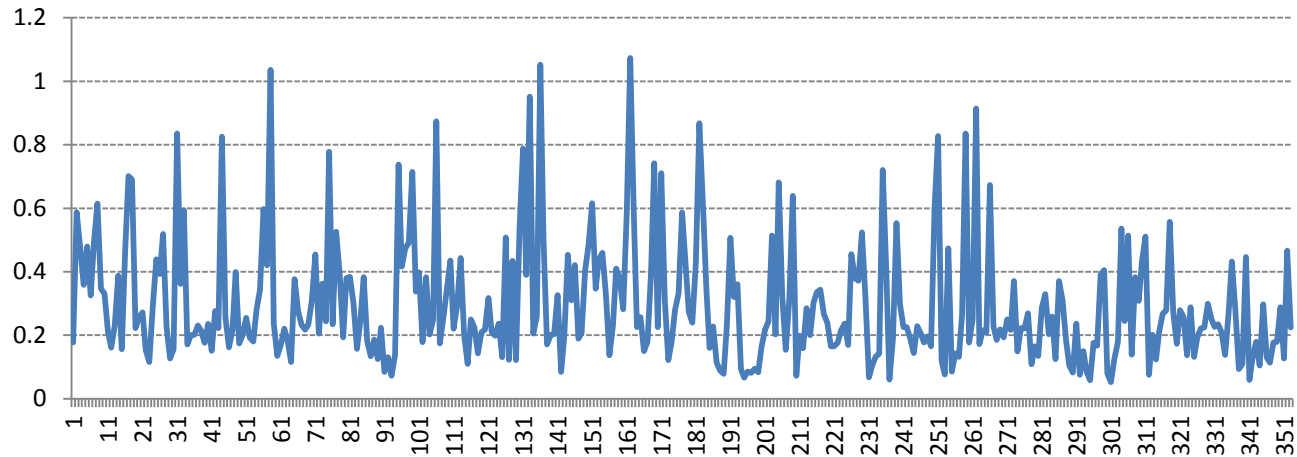
Seasonal component (12 seasons) of atmospheric NH_3+NH_4



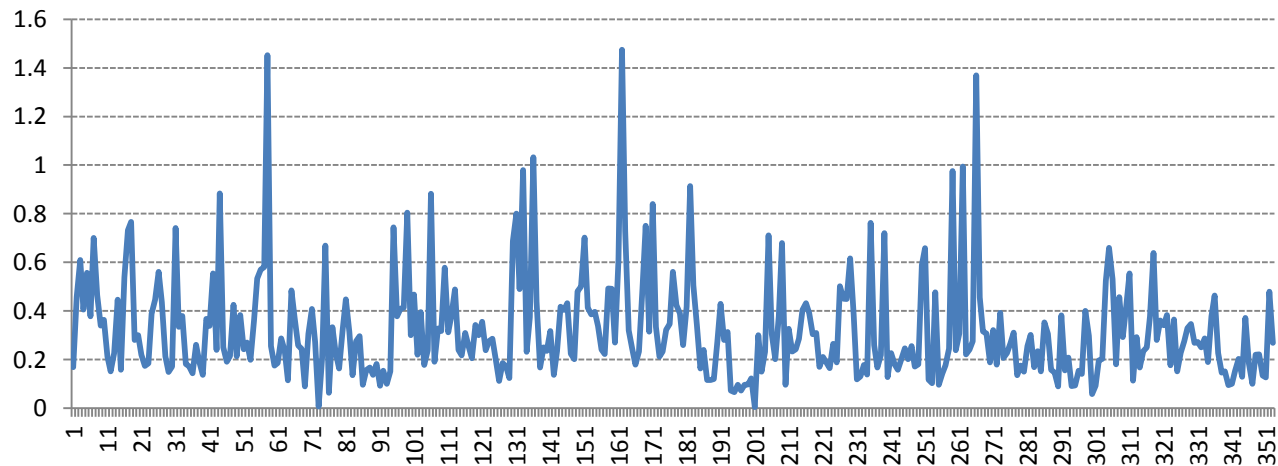
Large difference in spring

Next the time series were seasonalized for 52 seasons

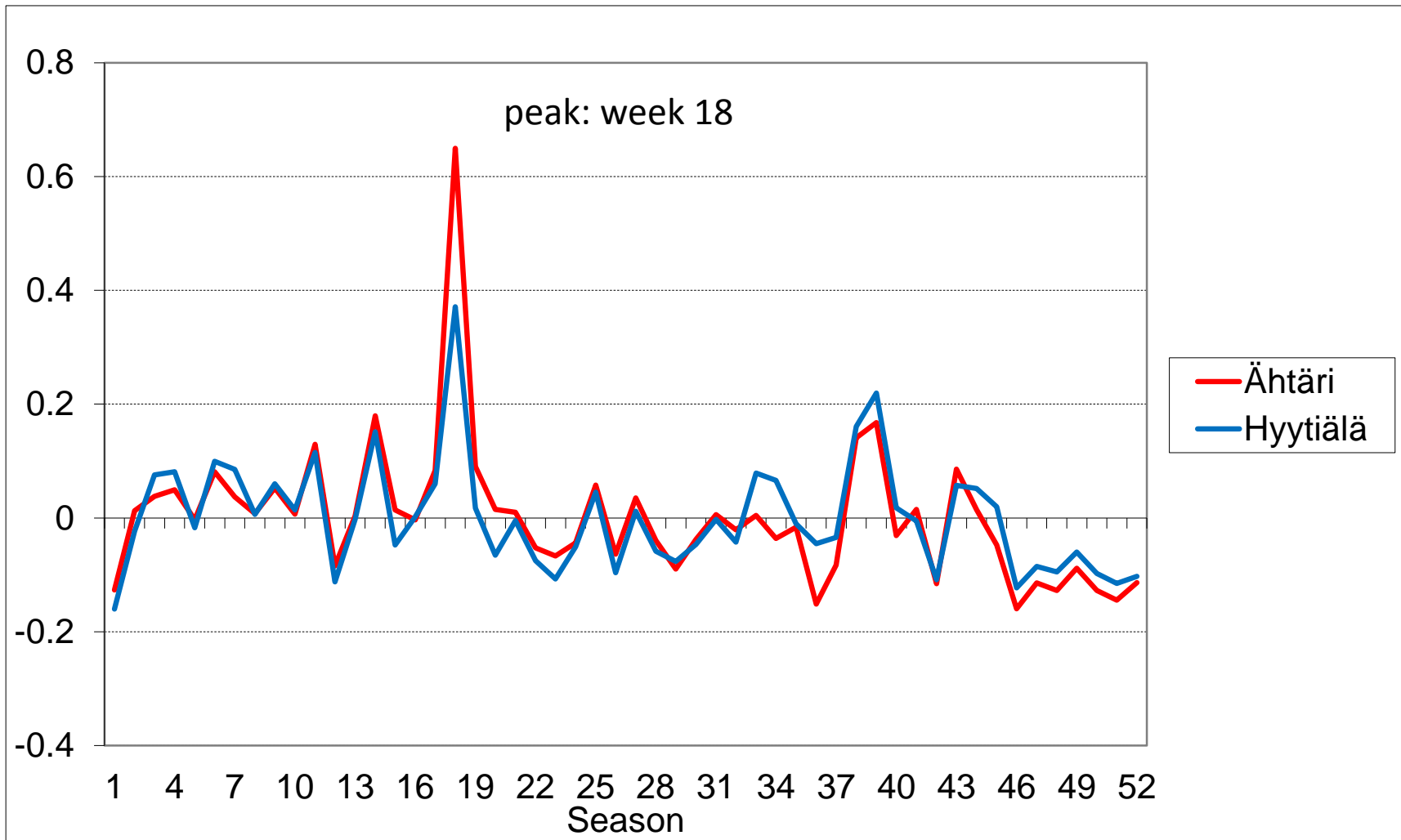
Hyytiälä



Ähtäri



$(\text{NH}_3 + \text{NH}_4)$ concentration in air, weekly mean values, $\mu\text{gN}/\text{m}^3$



Seasonal components (52 seasons) of atmospheric $\text{NH}_3 + \text{NH}_4$ concentration

- The largest difference between the stations in the seasonal cycle was in week 18 (late April or early May, depending on the year)
- In Finland, this is a time of high agricultural activity
- The spreading of manure (high NH_3 emissions) is permitted after 15 April on snow-free soil
- The snow melted 4 – 27 April at Ähtäri, and 28 March - 20 April at Hyytiälä in 2003–2009
- The emission estimate for NH_3 in 2000 at a 1 km x 1 km scale showed higher local emissions near Ähtäri than in the vicinity of Hyytiälä (Karvosenoja 2000).
- So, the general pattern of the 52 seasonal components is a result of LRT, and is rather similar at the two stations.
- The large difference during the spring is a result of different land-use patterns in the vicinity of the stations = local effect.

Conclusions

- Methods that take into account the seasonal behaviour and serial correlation in air quality time series can help to estimate the possible local contributions and establish their exact timing
- LRT is the most significant contributing factor in the time series
- At both sites, the high agricultural activity in spring is evident in the $(\text{NH}_3 + \text{NH}_4^+)$ data.
- With help of the dense seasonalization of the time series, the minimal part of the data affected can be removed and the remaining data used for further studies on a more regional level.