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## Trajectory-based source analysis

M. Hulkkonen et al.

# Trajectory-based source area analysis of atmospheric fine particles, SO<sub>2</sub>, NO<sub>x</sub> and O<sub>3</sub> for the SMEAR II station in Finland in 1996–2008

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

Source area analysis based on combining in situ measurements of trace gas or particle concentrations and back trajectories calculated for corresponding times has proven to be a valuable approach in atmospheric research; especially in investigating air pollution episodes, but also in e.g. tracing the source areas of air masses related to high vs. low concentrations of aerosol particles of different sizes at the receptor site. A statistical trajectory method used before by Sogacheva et al. (2005) was fine-tuned to take the presumable horizontal error in calculated trajectories into account, tested with SO<sub>2</sub> and validated by comparison against EMEP (European Monitoring and Evaluation Programme) emission data. In this work we apply the improved method for characterizing the source areas of atmospheric SO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub> and aerosol particles of different size modes from the perspective of a Finnish measurement station located in Hyytiälä (61°51' N, 24°17' E). Our method proved useful for qualitative source area analysis of measured trace compounds. We applied it to study trends and seasonal variation in atmospheric pollutant transport during 13 yr at the SMEAR II station.

## 1 Introduction

Statistical analysis of a large set of trajectories has been a popular tool for identifying the regions that serve as source areas of selected compounds and thus contribute to the concentrations measured at the receptor site (Stohl, 1996, 1998; Scheifinger and Kaiser, 2007). Different methods have been developed for the purpose of tracing back registered concentrations.

Ashbaugh et al. (1985) introduced a method that was later on named as Potential Source Contribution Function (PSCF). Similar concepts were developed by Vasconcelos et al. (1996) and Zhou et al. (2004). Instead of calculating conditional probabilities for high concentrations to occur as a result of certain air mass paths, actual concentration values for each grid cell are obtained with the Concentration Field (CF) method

## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



introduced by Seibert et al. (1994) and developed further by Stohl (1996). The results of both PSCF and CF methods can be interpreted as the distribution of either potential sources and sinks or the concentration of the compound. These methods have been applied in several studies run over Central Europe (e.g. Wotawa and Kröger, 1999; Kaiser et al., 2007; Apadula et al., 2003), most often with the intention to reproduce known emission fields. Trajectory statistical methods have been seen to construct characteristics of the emission fields with a good statistical significance (Kabashnikov et al., 2011).

Source area analysis done in previous studies for different trace gases and aerosol particles measured in Hyytiälä, Finland, has been based on classifying trajectories according to their origin (Kulmala et al., 2000; Hellén et al., 2004). By finding a set of trajectories with similar history and the average of concentration values measured at times corresponding to trajectory arrival times it has been possible to coarsely identify “clean” and “polluted” sectors from the perspective of Hyytiälä.

Source area analysis with the statistical trajectory method developed by Stohl (1996) has been done from the perspectives of other Finnish stations: to study the arrival of CO<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub>, black carbon and condensation nuclei to Pallas (Aalto et al., 2002); to find the areas contributing to O<sub>3</sub>, SO<sub>2</sub> and particle concentrations registered in Utö at the Baltic Sea (Engler et al., 2007) and to find the origins of sulphate, ammonium and sodium measured at Sevettijärvi (Virkkula et al., 1995). Since the flow climatology and background conditions are different at each site, the results can not be generalized to apply for Hyytiälä as well. Furthermore, we have a longer data set than in any previous study, which decreases the impact of exceptional years (e.g. years with strong forest fires) and episodes, and increases the statistical significance of the result. It is yet sensible to do some comparison between studies to possibly find features that are visible in all. Sogacheva et al. (2005) applied a similar method to aerosol particles of different size modes measured in Hyytiälä, but trace gas concentrations measured at SMEAR II have not been investigated with this kind of an approach.

**Trajectory-based  
source analysis**

M. Hulkkonen et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Trajectory-based  
source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The aim of this study is to distinguish areas that contribute the most and the least to the concentrations of  $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{O}_3$  and particulate matter measured in Hyytiälä. It is possible for an area to have an effect on concentrations elsewhere only if long-range transport takes place, i.e. when the area is passed by trajectories that are eventually received at the measurement site. The approach applied in our study allows us to recognize the relative contribution of each grid cell to the concentrations measured at Hyytiälä site. The product of our approach is visual and interpretable with one glance, but instead of absolute concentration values related to each sector we get the relative impact that different areas potentially have on measured values in Hyytiälä. When interpreting the results of source area analysis, it is thus necessary to ask, which factors have affected them the most. Some knowledge of existing point source emissions and flow dynamics is required.

Changes in the anthropogenic activities and natural phenomena can result in trends in the composition of the atmosphere. One aspect of this study is to find whether there has occurred strengthening or weakening in the contribution that different areas have on values observed in Hyytiälä over the 13 yr measurement period. Human influence and biogenic factors are also likely to cause monthly, seasonal and yearly variation in the results. We expect to detect these and discuss about the reasons behind them.

## 2 Materials and methods

For investigating the statistically significant source areas of different compounds from the perspective of a specific site we need continuous concentration measurements run over as long period as possible and reliably calculated back trajectories. The arrival height of trajectories should be close to the measurement height. Information about the site, its surroundings and the point sources of emissions also in a wider scale are crucial for drawing conclusions from the analysis results.

## 2.1 Atmospheric data from SMEAR II

SMEAR II (Station for Measuring Ecosystem-Atmosphere Interactions) measurement site is located in a rather homogenous Scots pine (*Pinus sylvestris*) stand on a flat terrain at the Hyytiälä Forestry Field Station of the University of Helsinki (61°51' N, 24°17' E, 181 m a.s.l.) 220 km north-west from Helsinki. The largest city near the SMEAR II station is Tampere, located about 60 km south-east from the measurement site and having about 200 000 inhabitants.

In an instrumented 73-m-tall mast there are monitors to measure several trace gas concentrations, temperature and wind speed profiles, the properties of solar and thermal radiation of the stand, and the fluxes between the canopy and atmosphere (Hari and Kulmala, 2005). Measurements have been run continuously since 1996. The data used in this study belongs to an extensive set of atmospheric measurements during 13 yr (1996–2008) at SMEAR II. SO<sub>2</sub> concentration is measured with a fluorescence analyzer (TEI 43 BS, Thermo Environmental), NO<sub>x</sub> (NO + NO<sub>2</sub>) concentration with a chemiluminescence analyser (TEI 42C TL, Thermo Environmental) and O<sub>3</sub> concentration with an ultraviolet light absorption analyser (TEI 49, Thermo Environmental); all at a height of 67.2 m above the mast base. Measured values are reported as 30 min means. Data coverage of the 13 yr measurement set is good, from 89 to 92%. Particle number size distributions are measured with a Differential Mobility Particle Sizer (DMPS) that consists of two DMA's and two CPC's and scans the size distribution of particles between 3–1000 nm in 10 min intervals (Aalto et al., 2001). Number concentrations for total particulate matter, nucleation mode (3–25 nm), Aitken mode (25–90 nm) and accumulation mode (90–1000 nm) particles are used.

## 2.2 Trajectories and data processing

HYSPLIT\_4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) trajectories (Draxler and Hess, 1998; Heinzerling, 2004) were calculated for an arrival height of 100 m above ground level with an hourly interval, 96 h back in time using NOAA

### Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



FNL-archive data (1° horizontal resolution, 13 pressure levels ) for 1998–2007 and NCEP/NCAR reanalysis data (2.5° horizontal resolution, 17 pressure levels) for years 1997 and 2008.

At each time step the measured concentration value is assigned to the grid cells (1° × 1°) along the corresponding back trajectory. Gas and particle concentrations were interpolated to the trajectory arrival times (on the hour) by using nearest neighbour interpolation. The horizontal uncertainty related to calculated HYSPLIT\_4 trajectories has been estimated to be 10–30 % of the distance travelled by the air parcel (15–30 % by Heinzerling, 2004; 10–20 % by Draxler and Hess, 1998). It is taken into account by assigning a weighted concentration value also to grid cells surrounding the trajectory path. Cells closer than 10 % of the trajectory travelling distance are given a concentration value weighted by 0.70 and those farther than 10 % but closer than 20 % of the travelled distance get a concentration weighted by 0.30. The choice of factors was made assuming a normally distributed probability of trajectory error. The method differs from the so called nine point filter suggested by Stohl (1996), where the first guess concentration field is followed by an iterative redistribution procedure to improve the spatial resolution.

Arithmetic or geometric mean of values accumulated to each grid cell is calculated. In the end the highest value in the fields under comparison is used for normalizing the values in each grid, the end result being a relative source contribution field (RSCF) that shows the relative contribution of each grid cell to the measured values monitored at the receptor site. In order to ensure the statistical significance of the result, values are calculated only, if a minimum number of trajectories, set to 10 in this study, crossed a grid cell. RSCF maps were produced for all compounds yearly, monthly over all years, and separately for periods 1996–1999 and 2005–2008.

### 2.2.1 Testing procedure of the trajectory method

The statistical trajectory method described in Sect. 2.2 was tested by applying it to sulphur dioxide concentrations measured in Hyytiälä. The method's sensitivity to how

## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Trajectory-based  
source analysis**

M. Hulkkonen et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

large an uncertainty of calculated back trajectories is taken into account proved to be quite big. Doubling the uncertainty, which meant giving weighted concentration values also to grid cells nearer to the trajectory path than 20–40 % of the travelled distance, dispelled all finer features from the concentration field. In that case the concentration field method becomes computationally a little heavier, while its output starts to resemble qualitatively the information got from a much coarser sector-wise approach applied by Kulmala et al. (2000). Assuming the trajectory uncertainty to be 5–10 % of the travelled distance, in turn, brought more features out of the concentration field making it very tempting to use, but since none of the HYSPLIT\_4 trajectory validation studies found in literature suggests so small an uncertainty, we ended up using 10–20 %.

The height of trajectories relative to the model estimate of mixing height was considered in order to address the fact that sources located in the surface layer do not affect the concentration found in the followed air parcel, when the height of its path exceeds mixing height. This is due to the zone between boundary layer and free troposphere, which in some cases forms a very stable capping inversion. The effect of the afore-described feature was tested in the model by leaving the grid cells, above which trajectory height exceeded mixing height, without a concentration value. This brought out finer features in the produced RSCF: it can be interpreted as a result of considering only the grid cells, the contribution of which to the values observed at the receptor site was real. Neglecting all grid cells along the trajectory after it rose above the mixing height for the first time, based on the assumption that the properties of the air parcel do not remain unchanged when visiting the free troposphere, led to almost identical results. This can be thought as a signal of the fact that it is the relatively strong sources located nearby that to some extent dictate the resulting RSCF. Taking only the lowest half of the mixing height into consideration led to no change in the RSCF which seems to show that the sources clearly in the lowest part of the boundary layer are dominant.

RSCF maps produced by the trajectory method with different assumptions were compared to EMEP emission data that is based on national monitoring and reporting (Vestreng and Klein, 2002), and also to concentration data modelled under the

EMEP program. The best correlation coefficient ( $R^2$ ) from Hyytiälä's perspective was found between the modelled concentration data and a RSCF produced by the trajectory method taking the height of trajectory relative to mixing height into account ( $R^2 = 0.11$ ). Combining data sets from different Finnish measurement stations (SMEAR I–III) did not bring improvements to the correlations with EMEP, although the number of trajectories was tripled and the subjective view of each station got less weight (Hulkkonen, 2010).

A conclusion was drawn that the existence of strong local or nearby sources may dominate the production of RSCF maps, which means that the interpretation should be done from the perspective of the receptor site and not as a model estimate of the real emission or source field – especially as long as the removal processes are not taken into account in a sophisticated manner. Although the absolute strength of the sources can not be detected, the method still allows us to see point sources also farther away from the receptor site, albeit qualitatively.

### 3 Site-specific features

The amount of hourly trajectories having passed each grid cell (within 50–75° N, 0–45° E) during 1996–2008 varies from 112 to 111 369. The dominant air mass flow direction from Hyytiälä's perspective is 220–310° (Fig. 1). The majority of trajectories received in Hyytiälä cross over the coast of Norway and the Scandis that have a cleansing effect on the air as a result of orographic precipitation. Also air masses crossing the Baltic Sea can be “cleaned” when encountering the coastline of Finland. After that, the significance of local sources is enhanced. Continental air can be expected to bring the highest concentrations of trace gases and particles for which wet deposition is an important sink.

The median wind speed along trajectories varies between 2.8–14 m s<sup>-1</sup>, with elevated values in winter months as a result of stronger temperature and pressure gradients. There is also significant variation in the flow speed along each trajectory: the

## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



difference between maximum and minimum flow speeds along a trajectory is as high as  $5.6\text{--}17\text{ m s}^{-1}$ , again with larger values in the winter. Varying flow speeds result in the variation of residence times over different areas and the possibility for short-lived compounds to travel long distances.

Also the altitude of the followed air parcel shows variation both along the trajectory and seasonally. When looking at the median of trajectories calculated for an arrival height of 100 m, the altitude increases steadily when receding from Hyytiälä and reaches approximately 350 m after 96 h. In winter months air parcels travel further and reach higher altitudes than in the summer: the maximum height achieved along a  $\sim 96$  h trajectory is approximately 5000 m in the winter and 1500 m in the summer. But in all seasons the majority of trajectories travel all the way under the altitude of 1000 m. The distance travelled by back trajectories in 96 h varies between 500–6000 km.

Hyytiälä can be considered as a background station in terms of air pollution. This becomes evident when looking at the median, minimum and maximum concentrations of atmospheric  $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{O}_3$  and particles of different size modes presented in Table 1. Seasonal variation exists (Lyubovtseva et al., 2005), but in general the background concentrations are low, and elevated values are typically registered in episodes. There are point sources of  $\text{SO}_2$ ,  $\text{NO}_x$  and particulate matter in the vicinity of the measurement station, for example UPM Kymmene paper mills approximately 80 km and 100 km away (visualized by Smart-SMEAR: Junninen et al., 2009). The  $\text{SO}_2$ - and  $\text{NO}_x$ -emissions from these, however, are three to four times smaller than those of the strongest point sources in Finland (e.g. Rautaruukki steelworks about 300 km away from Hyytiälä or powerplants in Helsinki, 230 km away). The highest values of Finnish point source emissions are of the order of  $4000\text{ t a}^{-1}$  for  $\text{SO}_x$ ,  $3200\text{ t a}^{-1}$  for  $\text{NO}_x$  and  $900\text{ t a}^{-1}$  for particulate matter ( $\text{PM}_{10}$ ) (E-PRTR – European Pollutant Release and Transfer Register, <http://prtr.ec.europa.eu/>). These values can be compared with those of a power plant located in northern Estonia, about 500 km away from Hyytiälä:  $47\,400\text{ t a}^{-1}$ ,  $8440\text{ t a}^{-1}$  and  $2500\text{ t a}^{-1}$  for  $\text{SO}_x$ ,  $\text{NO}_x$  and  $\text{PM}_{10}$ , respectively. St. Petersburg region with several point sources is approximately 700 km from Hyytiälä measurement site: if we assume

## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



an average flow speed of  $20 \text{ km h}^{-1}$ , an air parcel passing over St. Petersburg arrives in Hyytiälä in 35 h. This should be borne in mind together with the average atmospheric lifetimes of different compounds presented in Table 1. E.g. the amount of  $\text{NO}_x$  emitted in St. Petersburg would decline to  $1/e$ :th (assuming an exponential removal function) by the time it gets to the measurement site, since the travelling time approximately equals the atmospheric lifetime of  $\text{NO}_x$ .

## 4 Results and discussion

The produced RSCF maps are principally a representation of the combination of the flow dynamics relevant for Hyytiälä and the emission sources along air mass paths. The approach applied in this study diminishes the effect that different amounts of trajectories having crossed each grid cell might have on the calculation of the RSC-value, but if there is a very repetitive path crossing a grid cell with a strong emission source, the method gives erroneous weight to the grid cells along the trajectory farther from the actual source area. This has to be kept in mind when interpreting the RSCF maps.

The sectors used in trend analysis are presented in Fig. 2. They were chosen to represent homogeneously marine (A) and continental (C) areas, and the vicinity of Hyytiälä (B).

### 4.1 Source area analysis for aerosol particles of different size modes

The results for different size modes of aerosol particles are in line with previous studies (Kulmala et al., 2000; Sogacheva et al., 2005; Dal Maso et al., 2007), but reveal more detailed characteristics of the origins and pathways of air masses leading to observing high and low particle concentrations in Hyytiälä. Monthly, seasonal and annual variations, inter-modal differences and weakening or strengthening of different source areas over the measurement period of 13 yr are detected.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Trajectory-based  
source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



For the number concentration of nucleation mode particles ( $D_p = 3\text{--}25\text{ nm}$ ) the produced RSCF shows well outlined features that have seasonal variation (Fig. 3). Figure 3 gives also indication on the relative number concentration levels in different months. The frequency of new particle formation has been shown to peak in March–April and September (Dal Maso et al., 2007).

The source area features of nucleation mode particles remain similar over years, but a decrease in the levels of RSC-values can be detected: a decrease of  $-12\%$  in the median value in the marine sector A,  $-18\%$  in the Hyytiälä sector B and  $-30\%$  in the continental sector C between 1996–1999 and 2005–2008 (Table 2). This is in line with other studies, since nucleation mode particle concentrations have been detected to decrease in Europe due to decreased  $\text{SO}_2$  emissions (Hamed et al., 2010).

For measuring high nucleation mode particle number concentrations in Hyytiälä it is the air masses with north-western and Atlantic origin that are favourable. Clean arctic and marine air masses have been shown to be optimal for new particle formation in Hyytiälä (Dal Maso et al., 2007), while polluted continental air tends to prevent it due to a large pre-existing particle surface area (especially Aitken and accumulation mode particles) leading to high condensation and coagulation sinks and removal of nucleation precursor vapours. This can be deduced also from the RSCF maps produced for particles of different sizes. For example the dominant source sector for accumulation mode particles can indeed be found in the south-east indicating continental origin, which is the source sector of air masses connected to the lowest nucleation mode particle concentrations measured in Hyytiälä (comparing Figs. 3 and 4). Seasonal variation of RSCF for accumulation mode particles is shown in Fig. 4 and can be thought as a balance between winter time combustion related sources and summer time forest fires.

There is a slight decrease in accumulation mode particle sources in Table 2:  $-12\%$  over the Atlantic ocean (sector A) and  $-5\%$  over the continent (sector C).

In terms of total particle concentration, the source areas are not consistent from year to year. Marine and continental air masses alike are able to bring high concentrations.

This is visible in Fig. 5, where the yearly variation in the contribution of different areas to the total particle number concentrations measured in Hyytiälä is presented. For example years 2002 and 2003 with an exceptionally high fraction of days with new particle formation occurring stand out with the north-western sector having a strong contribution to the values measured in Hyytiälä. On the other hand, in the RSCF of 2006 the south-eastern corner clearly dominates as an indication of the forest fires that occurred in Russia that year (Riuttanen, 2009; Riuttanen et al., 2012; Saarikoski et al., 2007; Anttila et al., 2008).

A decrease in the total fine particle ( $D_p = 3\text{--}1000\text{ nm}$ ) concentration sources is detected at all three sectors. This is in line with previous studies of fine particle concentrations (e.g. Dal Maso et al., 2008).

A strong yearly variation applies also for the source areas of Aitken mode particles. It is both nucleation mode particles related to clean marine air masses and combustion related particles with continental origin that contribute to the number concentrations in Aitken mode. The twofold origin of air masses that is optimal for Aitken mode particles to occur in Hyytiälä is evident in the RSCF presented in Fig. 6, which shows the seasonal variation averaged over 13 yr. From April to September we observe elevated values throughout the field, which highlights the biogenic origin of Aitken mode particles. The north-west sector stands out, which possibly indicates ongoing nucleation and growth of particles as a result of condensing organic vapours along the way towards Hyytiälä, where we observe these grown particles instead of freshly nucleated ones throughout the summer. No change in the marine source between years 1996–1999 and 2005–2008 has been detected, but the continental source (sector C) has decreased by 10 % (Table 2).

Condensation sink (CS), the ability of an aerosol population to remove condensable vapour from the air, was calculated based on the particle size distribution information received from DMPS measurements in Hyytiälä. Air masses that lead to high condensation sink values are carrying a large particle surface area, typically dominated by particles belonging to the accumulation mode. Therefore, the studied source contribution

**Trajectory-based  
source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



fields of the CS have similar patterns as those for accumulation mode (Fig. 4).

## 4.2 Source area analysis for trace gases

### 4.2.1 SO<sub>2</sub>

A clear decrease in the SO<sub>2</sub> concentration level, presumably as a result of pollution control and the change in the East European industry in the late 1990's (Vestreng et al., 2007), can be detected when comparing periods 1996–1999 and 2005–2008. Changes of circa –50 % between the two periods are observed (Table 2).

Seasonal variation in the source areas of SO<sub>2</sub> is clearly visible in the RSCFs presented in Fig. 7: winter months from December to April are connected with the highest values as a result of higher demand for heating in the cold season, less photochemical loss of SO<sub>2</sub> and frequent inversions leading to the accumulation of concentrations.

The source area of the highest SO<sub>2</sub> concentrations measured in Hyytiälä is dominated by the south-east sector of the RSCF map. Clusters of anthropogenic emission sources in St. Petersburg, Baltic countries, Kola Peninsula and the south-east corner of the White Sea tend to stand out. Comparison against EMEP emission data (Hulkkonen, 2010) shows that the regions with the highest emissions are located in the south-west of the domain, but their relative contribution to the concentrations in Hyytiälä remains insignificant.

### 4.2.2 NO<sub>x</sub>

The importance of local sources in Southern Finland to measured NO<sub>x</sub> concentrations in Hyytiälä is seen in Fig. 8. NO<sub>x</sub> concentrations are the highest during winter months and early spring, because of combustion sources and weakness of the photochemical sink.

The seasonal variation of air mass origins connected with high NO<sub>x</sub> concentrations follows a very similar pattern as that for SO<sub>2</sub> described above. High concentrations in

## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Hyytiälä are more often than not related to air masses passing over St. Petersburg and the Baltic countries. The atmospheric lifetime of  $\text{NO}_x$  is typically of the order of 1 day, which means that the emission domain reliably seen by the method is smaller than shown in the RSCF figures. The areas with high anthropogenic  $\text{NO}_x$  emissions in St. Petersburg region and the northern Baltic are at the edge of this domain determined by the lifetime of  $\text{NO}_x$ , which explains the broadening “tail” of high values behind them from Hyytiälä’s perspective.

A decreasing trend in the general concentration level can be observed (Table 2: –25 % in sector A coming from the sea, –28 % in the vicinity of Hyytiälä and –15 % in the most polluted sector C), while the dominating sector remains very similarly delimited over the years.

### 4.2.3 $\text{O}_3$

In the case of ozone, there are no significant changes in its tropospheric concentrations over years from Hyytiälä’s perspective. The chemistry related to  $\text{O}_3$  might be too complicated for the trajectory method to capture the features of its source areas reliably, though. However, especially the monthly variation in the RSCF of  $\text{O}_3$  shows logical traits (Fig. 9). Air masses coming from marine areas (North-Atlantic and the Arctic Sea), where ozone deposition and the concentrations of ozone destroying pollutants are low, are connected with high  $\text{O}_3$  concentrations measured in Hyytiälä in the winter time. When approaching the summer, photolysis is gaining ground and there is a spring maximum of ambient ozone concentration. Also a shift of origin of air masses bringing high concentrations occurs between north-west and south-east, the latter sector being dominant in the summer and relatively stronger than the winter time source areas. Observed trends in ozone concentrations are minor. A slight increase in ozone sources in sectors A and B, and a slight decrease in the continental sector C have been detected between years 1996–1999 and 2005–2008 (Table 2).

## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 5 Conclusions

The relatively simple trajectory-based method applied in this study was evaluated with the conclusion that it can well be used for analysing the origins and paths of air masses related to high vs. low concentrations of atmospheric constituents monitored at the receptor site. If the height of trajectories relative to mixing height, relevant removal processes and the time spent in each grid cell by the air parcel were taken into account, we would approach the ability to produce fields that correspond to actual emission sources. The results obtained in this study disclose the contribution that different areas passed by air masses have on the values monitored in Hyytiälä, supporting earlier studies and reinforcing our conception of the regions that influence the measurement site in terms of different atmospheric constituents.

The method has been seen as a valuable tool for investigating source areas of transported atmospheric constituents. The improved RSCF method has already been used to study origins of aerosols with different optical properties (Virkkula et al., 2011). Possibilities for applications remain.

The origin of air masses that statistically bring high particle concentrations to Hyytiälä depends highly on which particle size mode is focused on. North-west dominates as the origin of air masses that relates to high nucleation mode particle concentrations, whereas air masses from the south-east bring high concentrations of combustion related accumulation mode particles. Particles belonging to Aitken mode get contribution from both directions. Seasonal variations in the RSC fields tell more about the nature of particles in each mode: for example RSCFs for Aitken mode particles show elevated values from April to September highlighting the biogenic origin, whereas for accumulation mode particles both winter time combustion and summer time forest fires can be deduced as factors defining the seasonal variation in the RSCFs. A general trend of weakening fine particle sources has been detected.

Combustion-related trace gases  $\text{SO}_2$  and  $\text{NO}_x$  are transported to Hyytiälä mainly from Eastern Europe, with winter months dominating. Local sources are also strong.

### Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Trajectory-based  
source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



A rather sharply outlined sector in the south-east has the strongest contribution to the high concentrations measured in Hyytiälä. This is presumably due to the fact that there are relatively strong emission sources in the industrial areas of northern Estonia and St. Petersburg, which are located so that for an air parcel passing over them, on average, it takes about 1–2 days to arrive in Hyytiälä. Being at the upper limit of the lifetime of the compounds in question, the travelling time from regions beyond the above-mentioned is so long as to make it impossible to see them as source areas from Hyytiälä's perspective. These regions are thus interpreted as areas passed by trajectories that eventually crossed also e.g. St. Petersburg and brought high concentrations to Hyytiälä. Furthermore, a decreasing trend of 25–50% in the general concentration levels of SO<sub>2</sub> and NO<sub>x</sub> between periods 1996–1999 and 2005–2008 is observed in the RSC fields.

The source areas of new nucleation mode particles have to be considered in the light of the processes that are producing these particles. The exact mechanism of atmospheric particle formation is yet unknown, but it has been established that sulphuric acid, resulting from the oxidation of SO<sub>2</sub> by photochemically produced hydroxyl radicals, is a major contributor to the formation process (Riipinen et al., 2007; Sipilä et al., 2010). Additionally, oxidised organic compound that are emitted from forested areas are probably also required to form observable particles (e.g. Paasonen et al., 2010). A critical inhibiting factor for particle formation is the pre-existing aerosol's ability to scavenge vapors and small particles from air is characterized by the condensation sink, CS (see e.g. Hyvönen et al., 2005; Dal Maso et al., 2007).

Studying the Relative Source Contribution Fields shows that SO<sub>2</sub> and CS source contributions seem to originate from roughly the same geographical areas, namely continental (Eastern) Europe (Figs. 7 and 4), possibly even from partly the same sources. The air masses contributing to higher nucleation mode particle concentrations, however, seem to originate from a different area, e.g. the Scandinavian sparsely populated areas and the Arctic Ocean. This result is similar to results obtained in previous studies (e.g. Sogacheva et al., 2008; Nilsson et al., 2001). The comparison of the different

contributing process indicators (CS for loss processes, SO<sub>2</sub> for sources) also supports the idea that for the Hyytiälä station, particle formation is controlled by the particle loss processes rather than the particle source processes (as already suggested by Dal Maso et al., 2007). This might have consequences for aerosol formation in the future, if air pollution regulations lead to reductions in either (or both) SO<sub>2</sub> and particulate matter and thus CS.

The diversity of processes affecting tropospheric ozone concentrations is a challenge for the simple source area analysis conducted in this study. The lifetime of ozone has a strong seasonal variation mainly due to a varying photolysis rate, which affects its long-range transport. The origin of air masses that bring high concentrations of O<sub>3</sub> to Hyytiälä shifts between north-west and south-east, the latter sector being dominant in the summer and relatively stronger than the winter time source areas that are congruent with areas characterized by low concentrations of ozone destroying pollutants. Unlike for SO<sub>2</sub> and NO<sub>x</sub>, no significant changes in the general concentration level of tropospheric O<sub>3</sub> is observed over the 13 yr measurement period in Hyytiälä.

Some difficulties related to the method and the interpretation of the results remain – especially when investigating compounds with varying lifetimes and those with strong sources in the proximity of the measurement site. Also the reliability of HYSPLIT trajectories in North-European context should be studied in more detail. Potential emission sensitivity or so-called footprint calculated by e.g. FLEXPART (Stohl et al., 1998) could lead to a better description of the actual source areas than single trajectories. The extent of the data set used in this study (9000–113 000 trajectories per figure) is, however, good enough to produce results with statistical significance – and with less computational capacity needed.

*Acknowledgements.* This research was supported by the Academy of Finland Center of Excellence program (project number 1118615).

**Trajectory-based  
source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## References

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## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Trajectory-based  
source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Trajectory-based  
source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Trajectory-based source analysis**

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Trajectory-based  
source analysis

M. Hulkkonen et al.

**Table 1.** Investigated compounds, their typical sources, sinks and estimated atmospheric life-times (according to Seinfeld and Pandis (1998) for trace gases and Lauer and Hendricks (2006) for particles). Median, minimum (median of values < 1-percentile limit) and maximum (median of values > 99-percentile limit) concentrations registered in Hyytiälä 1996–2008.

Compound	$\tau$	sources	sinks	Hyytiälä measurement statistics 1996–2008		
				median	minimum	maximum
NO <sub>x</sub> [ppb]	1.5 d	Fossil fuel (combustion); Biomass burning; Soils; lightning; aircrafts	Photochemistry; Oxidation to HNO <sub>3</sub> and PAN; Dry deposition	1.33	0.15	10.0
SO <sub>2</sub> [ppb]	2 d	Fossil fuel (combustion + industry); Biomass burning; Volcanoes; Oxidation of DMS	Dry deposition; Wet deposition; Chemistry (reactions with OH radical)	0.18	< 0.01	4.4
O <sub>3</sub> [ppb]	8 d (summer) – 100 d (winter)	In situ chem. prod.; Transport from the stratosphere; Interhemispheric transport	In situ chem. loss (photochem.); dry deposition	32.3	7.00	62.0
Particle number concentration ( $N$ ) ( $D_p = 3\text{--}25\text{ nm}$ ) [cm <sup>-3</sup> ]	< 1 d	Nucleation	Brownian coagulation; Condensational growth	238.9	5.7	8078
$N$ ( $D_p = 25\text{--}90\text{ nm}$ ) [cm <sup>-3</sup> ]	0.5–2 d	Condensational growth of nucleation mode particles; Emissions	Inter- and intramodal coagulation; Dry deposition	755.9	63.9	5412
$N$ ( $D_p = 90\text{--}1000\text{ nm}$ ) [cm <sup>-3</sup> ]	4–6 d	Traffic; Growth of Aitken mode particles	Wet deposition; Intramodal coagulation	445.5	36.4	2972
$N$ ( $D_p = 3\text{--}1000\text{ nm}$ ) [cm <sup>-3</sup> ]	5–7 d	–	–	1785	211	11 980

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



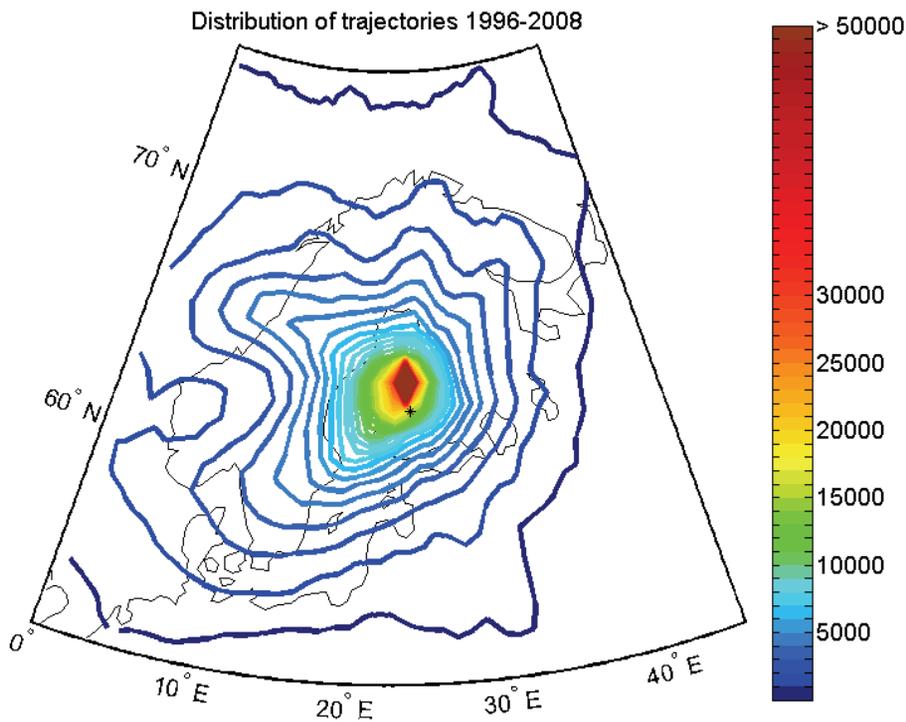
## Trajectory-based source analysis

M. Hulkkonen et al.

**Table 2.** Change in median RSC-values of atmospheric constituents in sectors represented in Fig. 2 between years 1996–1999 and 2005–2008.

	sector A	sector B	sector C
$N_{D=3-25\text{nm}}$	–12 %	–18 %	–30 %
$N_{D=25-90\text{nm}}$	–0 %	–13 %	–10 %
$N_{D=90-1000\text{nm}}$	–12 %	–9 %	–5 %
$N_{D=3-1000\text{nm}}$	–4 %	–15 %	–12 %
$\text{SO}_2$	–53 %	–54 %	–50 %
$\text{NO}_x$	–25 %	–28 %	–15 %
$\text{O}_3$	2 %	3 %	–7 %

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[I◀](#)
[▶I](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

**Fig. 1.** The distribution of trajectories 1996–2008. The amount of hourly trajectories having passed each grid cell is shown with coloured contour lines.

## Trajectory-based source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

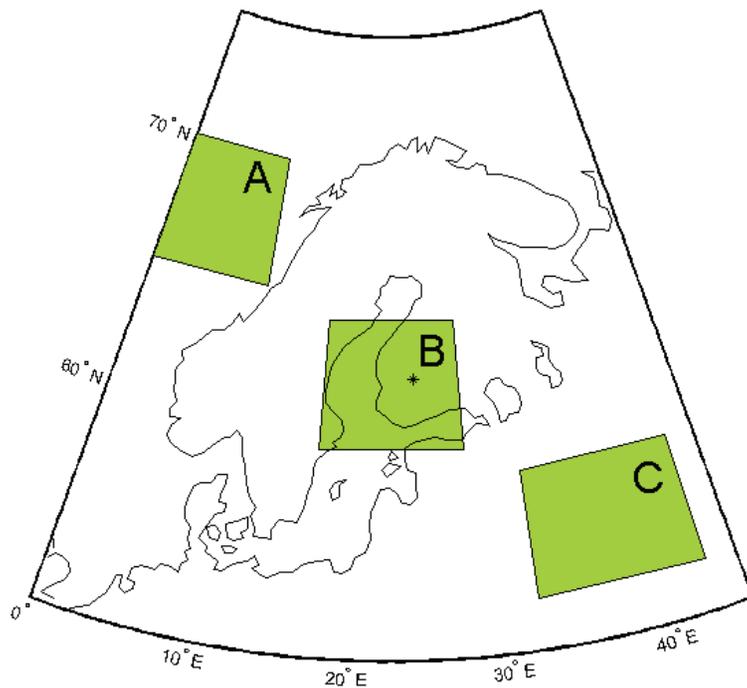
Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





**Fig. 2.** Three sectors were selected to study concentration trends. Sector A represents marine environment, sector B the vicinity of Hyytiälä measurement station and sector C continental environment.

**Trajectory-based source analysis**

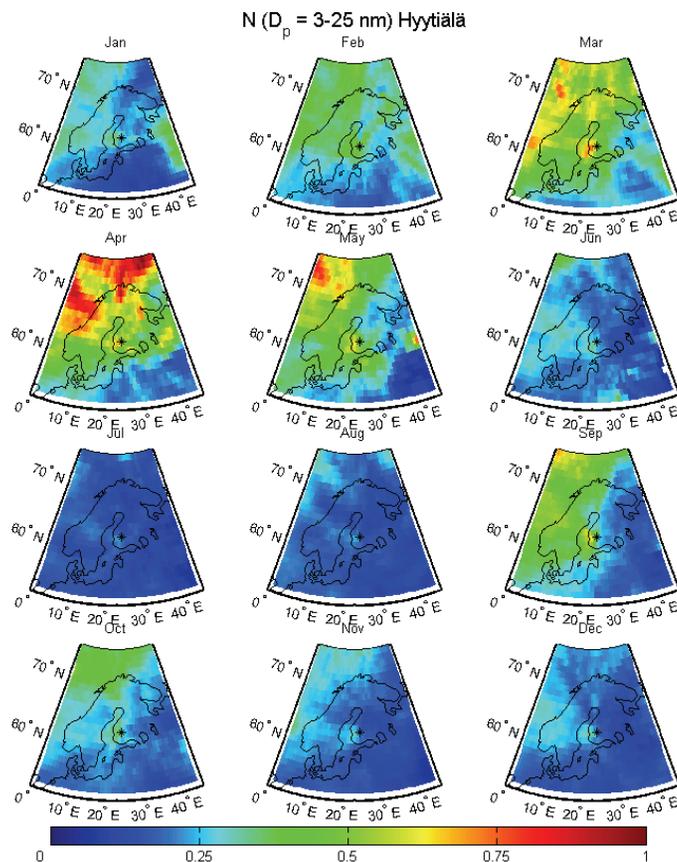
M. Hulkkonen et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



Trajectory-based  
source analysis

M. Hulkkonen et al.



**Fig. 3.** Monthly Relative Source Contribution Fields produced for the number concentration of nucleation mode particles ( $D_p = 3\text{--}25 \text{ nm}$ ) measured in Hyytiälä (marked with a star) 1996–2008. Higher values correspond to source areas leading to high concentrations measured at Hyytiälä.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

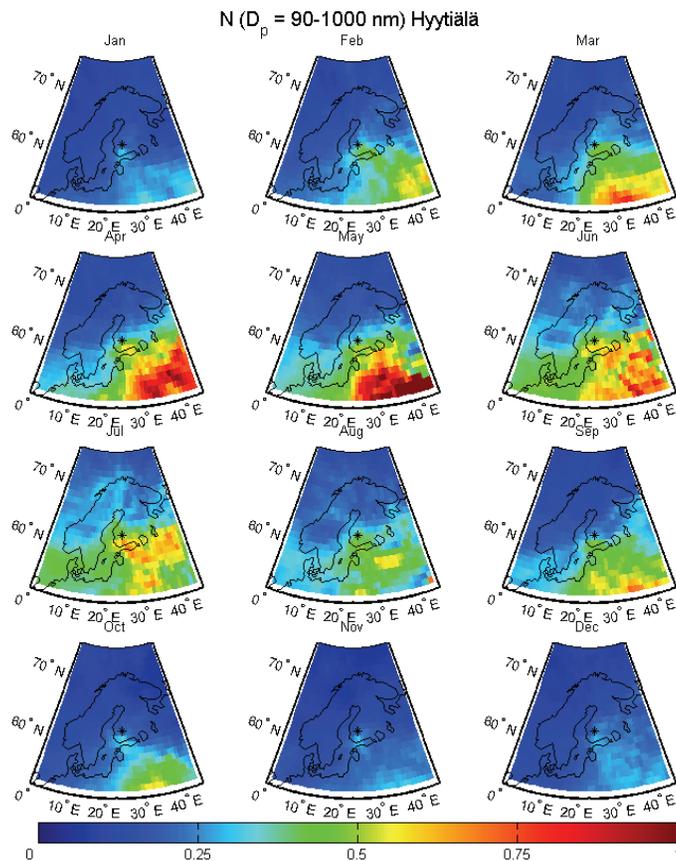
Printer-friendly Version

Interactive Discussion



Trajectory-based  
source analysis

M. Hulkkonen et al.



**Fig. 4.** Monthly Relative Source Contribution Fields produced for the number concentration of accumulation mode particles ( $D_p = 90\text{--}1000 \text{ nm}$ ) measured in Hyytiälä (marked with a star) 1996–2008. Higher values correspond to source areas leading to high concentrations measured at Hyytiälä.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Trajectory-based  
source analysis

M. Hulkkonen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

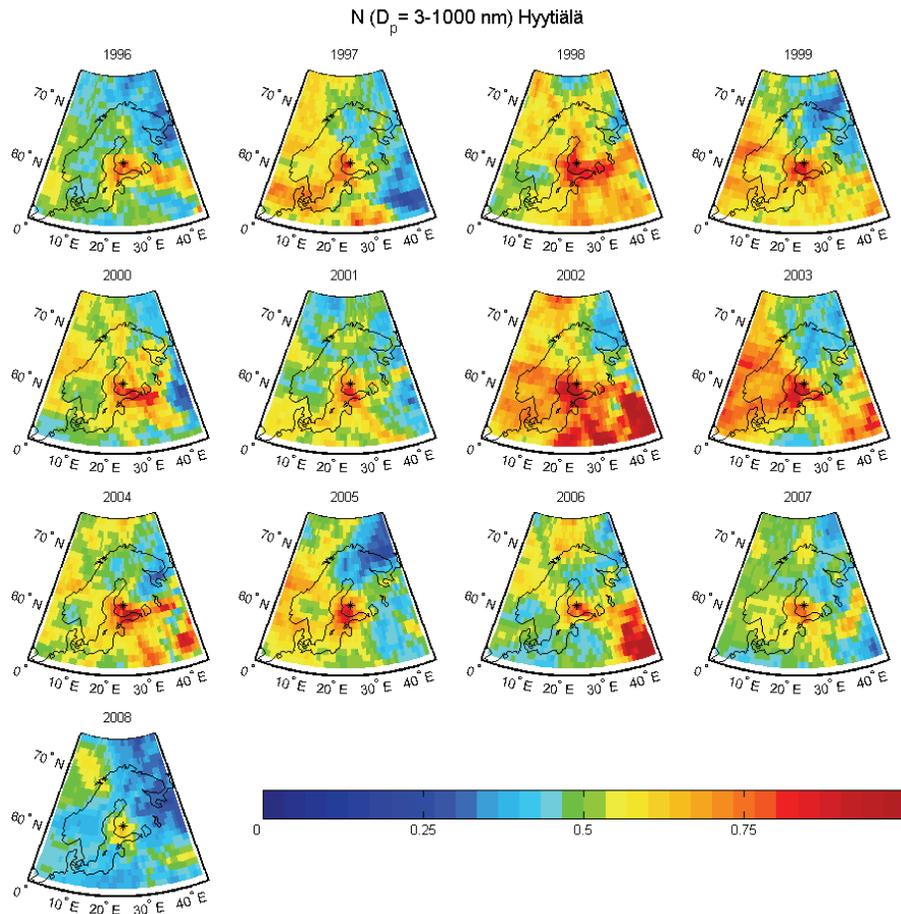
Back

Close

Full Screen / Esc

Printer-friendly Version

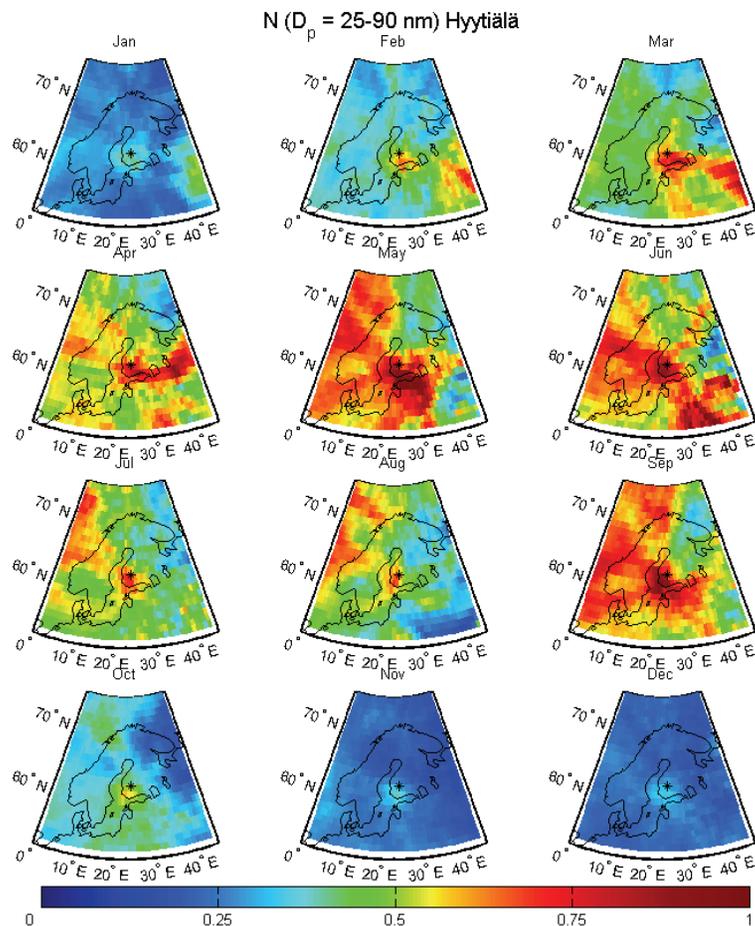
Interactive Discussion



**Fig. 5.** Yearly Relative Source Contribution Fields produced for the total number concentration of particles ( $D_p = 3-1000 \text{ nm}$ ) measured in Hyytiälä (marked with a star) 1996–2008. Higher values correspond to source areas leading to high concentrations measured at Hyytiälä.

Trajectory-based  
source analysis

M. Hulkkonen et al.



**Fig. 6.** Monthly Relative Source Contribution Fields produced for the number concentration of Aitken mode particles ( $D_p = 25-90 \text{ nm}$ ) measured in Hyytiälä (marked with a star) 1996–2008. Higher values correspond to source areas leading to high concentrations measured at Hyytiälä.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

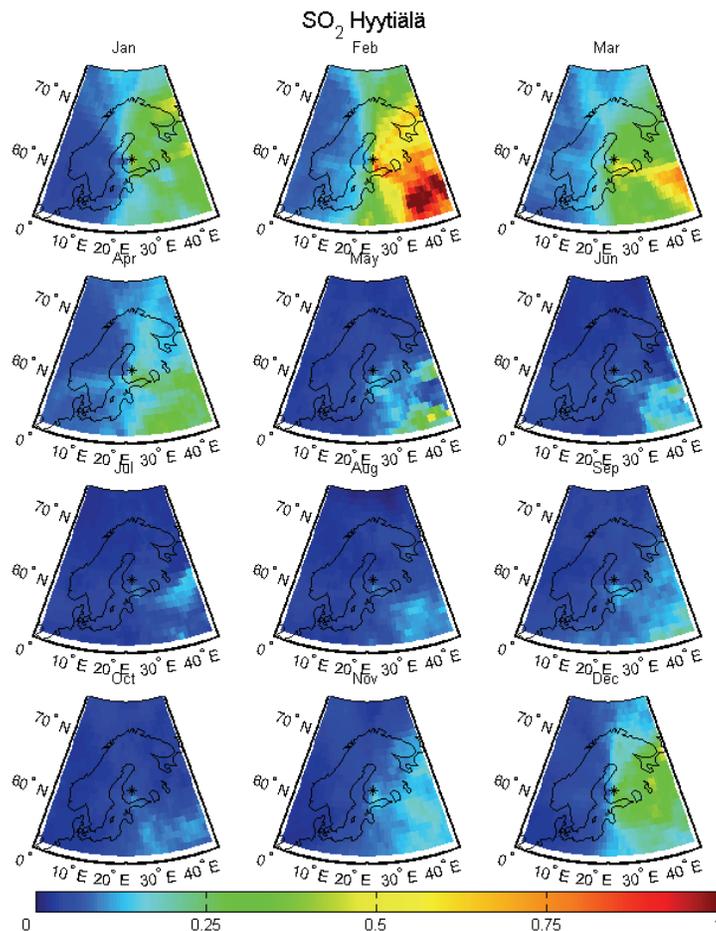
Printer-friendly Version

Interactive Discussion



Trajectory-based  
source analysis

M. Hulkkonen et al.



**Fig. 7.** Monthly Relative Source Contribution Fields for SO<sub>2</sub> concentrations measured in Hyttiälä (marked with a star) 1996–2008. Higher values correspond to source areas leading to high concentrations measured at Hyttiälä.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

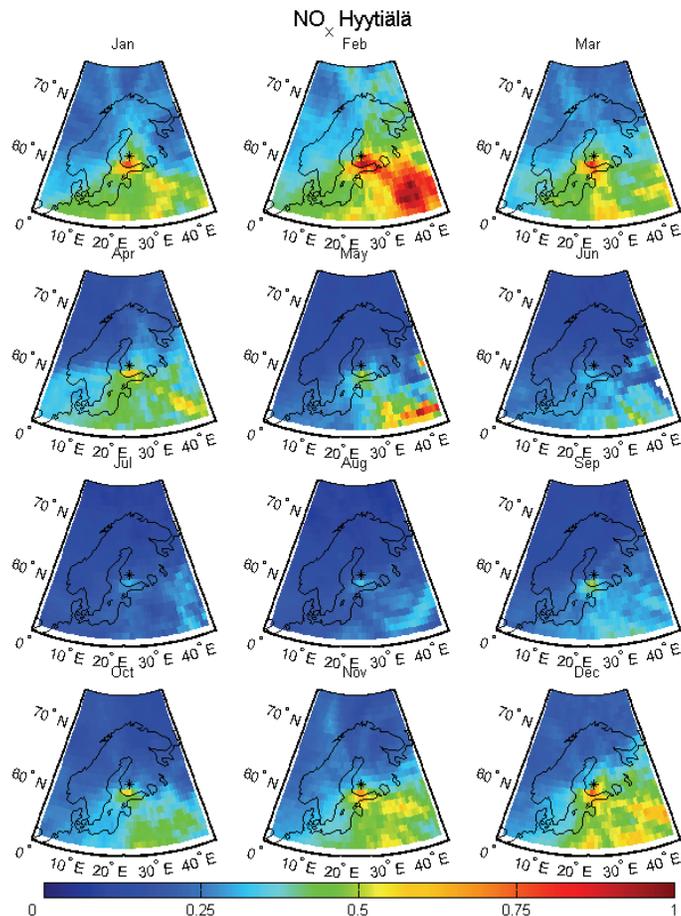
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**Fig. 8.** Monthly Relative Source Contribution Fields for NO<sub>x</sub> concentrations measured in Hyytiälä (marked with a star) 1996–2008. Higher values correspond to source areas leading to high concentrations measured at Hyytiälä.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

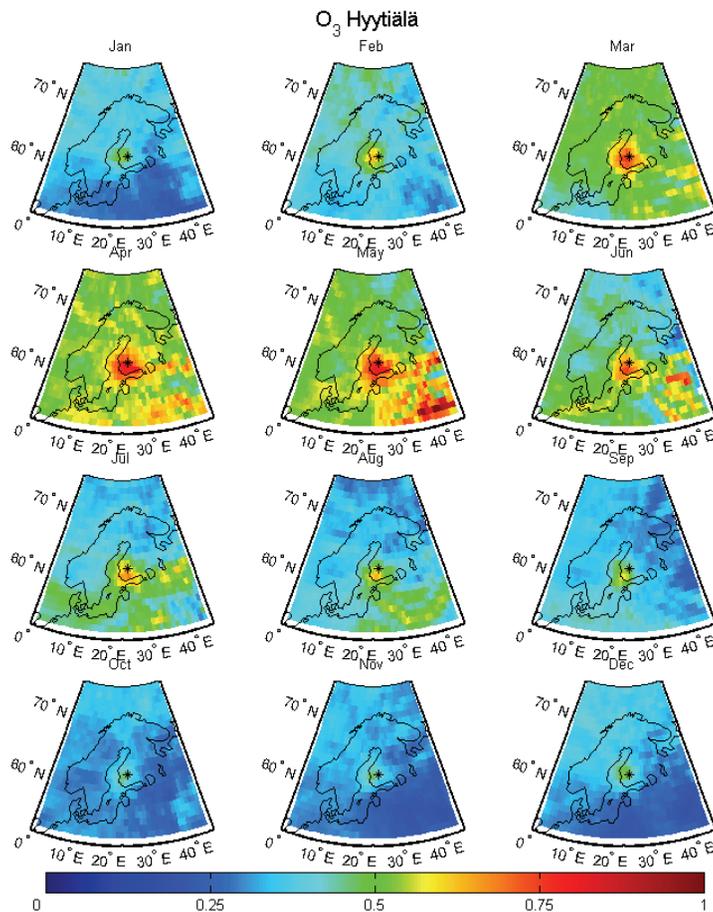
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**Fig. 9.** Monthly Relative Source Contribution Fields for O<sub>3</sub> concentrations measured in Hyytiälä (marked with a star) 1996–2008. Higher values correspond to source areas leading to high concentrations measured at Hyytiälä.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

