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Application of trajectory clustering for determining the source regions of secondary inorganic aerosols measured at K-puszta background monitoring station, Hungary

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Abstract Understanding the formation process of atmospheric particles is vital because of the significant impact of particulate matter on human health and climate change. Atmospheric particles can be formed by nucleation process via a number of different mechanisms, such as binary nucleation (involving H_2SO_4 and water vapour), ternary nucleation (involving NH_3, H_2SO_4 and water vapour) and ion-induced nucleation for charged particles, depending on the environmental conditions. Particle formation increases the total number concentration of ambient submicron particles and contributes thereby to climate forcing. The transformation processes of new particle formation (NPF) and secondary organic aerosol have been studied. It was found that gaseous sulphuric acid, ammonia, and organic compounds are important precursors to NPF events and H_2SO_4-NH_3-H_2O ternary nucleation is one of the important mechanisms. Using cluster analysis on the backward trajectories makes it possible to identify the most relevant types of air mass transport routes, and the directions from where precursor gases are transported. The influence of synoptic-scale atmospheric transport patterns on observed levels of sulphate, nitrate and ammonium has been examined.

1. Introduction

Atmospheric particulate matter (PM) can have both a natural and an anthropogenic origin that influences its composition and size. The effect of local meteorological conditions and in addition, long-range transport can have a significant influence on PM concentration levels recorded at a specific site (Abdalmogith and Harrison, 2005), but this is not currently well documented in many European geographical areas. Secondary inorganic aerosols (SIA) – SO_4^{2-}, NO_3^-, NH_4^+ – constitute a dominant part of particulate matter in Europe. They are called secondary because they are not emitted directly into the atmosphere but produced as a result of chemical reactions concerning sulphur dioxides, nitrogen oxides and ammonia. These gases are emitted by transport, industry and agriculture. During the last decades their emissions decreased considerably in Europe and this resulted in lower ambient concentrations for SIA.
New particle formation and subsequent growth of atmospheric particles have been observed in different environments: from sub-arctic Lapland and remote boreal forest to urban and suburban environments. Results showed that H_2SO_4 is the key compound affecting the NPF. However, before NPF, sulphuric acid clusters have to be

stabilized by other compounds. The most proper candidates could be amines, ammonia and extremely low volatile organics (Tao, 2016).
Combining advanced particle formation theories with cluster and backward trajectory analyses contributes to better understanding of the relevant atmospheric processes, and provides a useful tool for preparing effective air quality management strategies.

2. Methods

K-puszta is a background monitoring station, located in a clearing in a mixed forest on the Hungarian Great Plain in the middle of the Carpathian Basin. The nearest big city is Kecskemét located about 20 km to the southeast, opposite to the prevailing wind direction. In the direction of the prevailing wind being northwesterly the nearest anthropogenic source is Budapest the capital of Hungary located 80 km from the station, The Hungarian Meteorological Service performs the measurement of gaseous components and the analysis of precipitation, furthermore provides data for EMEP and WMO-GAW networks. The evaluated data are from K-puszta.
The commonly used Hybrid-Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015) developed by the National Oceanic and Atmospheric Administration's Air Resources Laboratory was applied. HYSPLIT uses archived 3-dimensional meteorological fields generated from observations and short-term forecasts. Three-day, hourly back trajectories for January 2007 to December 2015, arriving at the sampling location K-puszta at 1200 UTC and 800 m above ground level were calculated.
Trajectory clustering is a time series clustering problem. Time series clustering methods can be categorized into three main categories: feature-based, model-based, and raw-data-based (Liao, 2005). In the feature based methods, time series data are converted to static data and traditional clustering techniques, such as k-means, fuzzy c-means, and hierarchical clustering methods, are applied to the static data. Effectiveness of the feature-based methods highly depends on the effectiveness of two processes: conversion of the time series to a static value and clustering the static values. In model-based methods, a probabilistic model is assumed for the time series data, and the time series data is evaluated by its fitness degree to the applied model. The nature of the trajectory clustering problem is not suitable for model-based approaches, especially when the clusters are unknown. In raw-data-based methods time series data are given to the model as they are, without reducing to several features. Each time series data from the beginning to the end are considered as a sample for the model, and clustering is performed based on the output of the model. Several different raw-data-based methods exist in the literature, such as Dynamic Time Warping (DTW), Self Organizing Map (SOM) and various similarity/dissimilarity measures (Karaca, and Camci, 2010). Among the mentioned methods, the traditional clustering, k-means technique has been selected in this work. Nevertheless it has to be recognized that the k-means cluster analysis is sensitive to the selection of the initial cluster centers.

3. Results

The influence of synoptic-scale atmospheric transport patterns on observed levels of sulphate, nitrate and ammonium has been analysed. First the cluster centers had to be determined. The trajectory with the smallest total distance from the cluster center was is assigned to the cluster. Every cluster center is the arithmetic mean of all members of a cluster. Fig. 1. shows the finally specified cluster centers which are used for the examination of SIA origins. It can be seen that there are 8 dominant paths of air masses reaching Hungary.

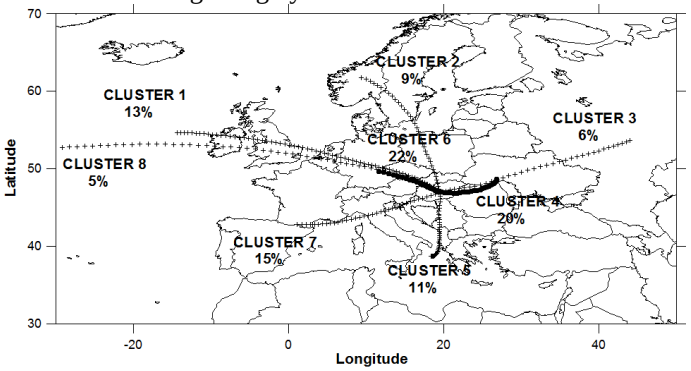

Fig.1. Cluster centers for the time interval 2007-2015

Annually, the most frequent patterns of the clusters coincided with cluster 6 and cluster 4, representing 22% and 20% of the days, respectively (see Fig 1.). These directions represent the main continental source areas of the precursor gases of SIA in east and west. According to the results, three potential source areas were identified as having important contributions to SIA in Hungary (see Fig. 2). Trajectory class mean NH_4^+ concentration ranged from 0.86 μgm^{-3} to 1.69 μgm^{-3}. The highest concentrations were strongly associated with air originated from eastern Europe (cluster 4) and a slow marine flow (cluster 5). The highest (2.41 μgm^{-3}) and lowest (1.7 μgm^{-3}) nitrate concentrations at K-puszta were associated with cluster 5 (southern) and cluster 3 (from Russia), respectively, whereas the highest (4.28 μgm^{-3}) and lowest (1.57 μgm^{-3}) sulphate concentrations were associated with the same clusters as the ammonium concentrations (see Fig. 2). Based on the sulphate and ammonium concentration ratios we can assume that sulphate mainly exist as $(NH_4)_2SO_4$ in K-puszta. Relatively high SIA concentrations were associated with eastern, southern and western pathways (cluster 4, 5 and 6). Probably these three continental paths were responsible for picking up air pollution over the continent of Europe and transporting them to the K-puszta station Malcolm et al. (2000) concluded that the concentrations are highest when air parcels pass over the European source region. These trajectories have short transport patterns, indicative of slow-motion air masses and represent the effect of continental air masses passed over countries, where industries are concentrated.

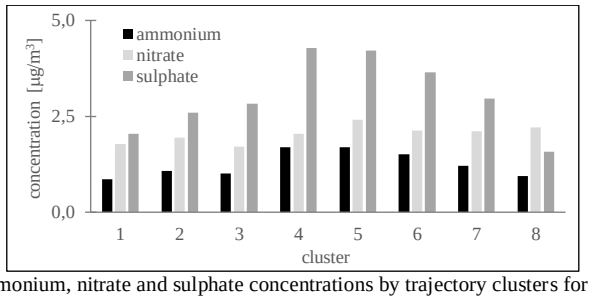

Fig.2. Mean ammonium, nitrate and sulphate concentrations by trajectory clusters for 2007-2015.

4. Conclusion

In this study, air mass backward trajectory cluster analysis was used to investigate the transport pathways and potential source of SIA in K-puszta monitoring station. Eight clusters were generated from backward trajectory cluster analysis for the period 2007-2015. Among them, two air mass trajectories originated from the continental region with a total occurrence frequency of 41% of the SIA concentration at K-puszta. The largest daily contribution to annual SIA concentration was observed for the continental clusters (clusters 4,5 and 6). Based on our preliminary results we can conclude, that the main SIA sources are in the continental regions in Europe.

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