Investigation of long-term changes of air pollutants concentrations in North-East Asia including Russia using EANET monitoring data

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Abstract

Permanent air quality monitoring networks provide valuable continuous measurement data. An interesting and important task is to analyze entire observational records revealing presence of monotonic long-term changes in air pollutant concentrations in the atmosphere. In this study we use EANET monitoring data on several major air pollutants for qualitative and quantitative assessment of trends at rural and remote stations in the North-East Asia region covered by EANET. Trends on the entire observational records as well as on selected seasonal subsets were estimated. For about a half of records considered we infer presence of statistically significant trends, whose spatial distribution we analyze further. Together with this trend assessment the comparison of mean air pollution levels at the North-East Asian (EANET) and European stations (from the European Monitoring and Evaluation Programme (EMEP)) is given. Statistical comparison of mean concentrations from the stations with similar environmental conditions was performed as well as comparison of seasonal trends.

Key words: EANET, Regional air pollution, Trend analysis

1. Introduction

One of the major aims of permanent monitoring networks is detection of long term changes and their similarity or difference over whole territory of interest. In addition, it is interesting to find interrelation between the changes in regions.

Long term changes of pollutants burden may be caused by different reasons. One of them is emission changes in the area affecting the station. This extent of this area can be large taking into account long range presence of one or many of them. Further investigations (e.g. modelling of long-range pollution transport) should be used to quantify these.
Trend assessment becomes more and more important taking into account changes in emissions and transport of pollutants in the atmosphere. For example, such work was done by European Monitoring and Evaluation Programme (EMEP) (http://www.nilu.no/projects/ccc/).

To obtain a reliable trend assessment, long-term high-quality monitoring data is needed. The minimum length of observational record required is determined by the specific questions and goals of the investigation. By now EANET provides fourteen years of continuous monitoring data that, as we show below, may be successfully used to obtain consistent trend estimates.

Annual and monthly means of pollutant concentrations observed during EANET monitoring period are available via annual EANET Data Reports (EANET. 2015). This information lets one to grasp the overall variance in annual/monthly averages, however it is not well-suited for a consistent trend assessment. There are several reasons for that: (1) Statistically significant presence of monotonic increase or decrease of pollutant concentrations in the entire observational record may be very sensitive to statistical outliers, which detection is determined by the total sample size and data distribution. The latter substantially differ for the annual/monthly means and the source “raw” data. (2) Most of the pollutants have characteristic atmospheric lifetimes shorter than a few months, therefore their seasonal abundances are determined by different superposition of the relevant processes involved. Consequently, trends estimated for the seasonally selected data exhibit changes caused by these (dominating) processes, often in contrast to the entire observational record. In this study, we address the above mentioned problems by scrutinizing raw measurement data for the entire observational periods, establishing proper outlier detection criteria and dissecting the data according to each geographical region, season and even each month.

Another task set by Network Center was comparison of pollution levels at EANET stations and European stations of EMEP network.

2. Methods

In this paper non-urban sites in North-East part of EANET region. Due to lack of measurement data at rural and remote sites in China we used monitoring data from stations located in Japan, Korea, Mongolia and Russia only. Figure 1 illustrates stations whose monitoring data we considered.

We use filterpack measurement data of air concentrations of gaseous and aerosol species (SO₂, NH₃, HNO₃, HCl, SO₄²⁻, NO₃⁻, Cl⁻, NH₄⁺, Na⁺, K⁺, Mg²⁺, Ca²⁺). A typical EANET biweekly sampling frequency yields approximately six points for each season and about two-dozen of points per year, respectively. Period of consideration was from 2000 to 2013, i.e. from the beginning of regular operation of the network.
Considered EANET stations

Figure 1. Rural/remote stations in North-East Asia in this work: Japan (Rishiri, Ochiichi, Tappi, Sado-Seki, Oki, Hapo, Ijira, Yusuhara, Hedo, Ogasawara), Korea (Kanghwa, Imsil, Cheju), Mongolia (Terelj), Russia (Mondy, Listvyanka, Primorskaya). Rural stations a presented in green color, remote stations are presented in blue.

Time series were analyzed for presence of statistical outliers whose appearance is caused by measurement/data processing errors, exceptional local pollution events and meteorological conditions, or unidentified reasons. Filtered time series were analyzed for statistically significant presence of trends. We considered entire observational records and also divided them into four seasons and analyzed those seasons separately. To assess trends we used median regression (MR), which is essentially the linear trend separating the halves of the given distribution (Koenker and Bassett, 1978). Trend was considered as statistically significant if according confidence intervals didn’t include zero and not significant otherwise trends.

To compare monitoring data in EANET and EMEP networks the several EMEP stations were chosen to be similar rural and background sites in accordance with EANET classification (Technical Manual for Air Concentration Monitoring in East Asia). The selected stations for comparison are: Nord (Greenland), Bredkalen (Sweden), Oulanka (Finland), Svratouch (Czech Republic), Leba and Śnieżka (Poland), Zingst (Germany). We used filterpack measurement data of these stations downloaded from Internet assessed data base of World Data Centre of Aerosols - EBAS (http://ebas.nilu.no/). In line with adopted sampling protocol EMEP sites provide one-day sampling filterpack data (EMEP Manual, http://www.nilu.no/projects/ccc/manual/index.html). Data used for comparison from mentioned stations cover mostly the same period of 2000 to 2012.

We used Student’s two sample t-test to investigate whether mean concentrations are equal or not and after that which concentrations (EMEP or EANET) are statistically significant higher. It is necessary to note that for EANET stations we have two-week mean concentration for one sample while for EMEP it is one day mean concentration for one sample. Moreover the periods of data at EANET and EMEP stations for comparison are not fully equal but the first of them (EANET data) usually contents the second one. The comparison is the most interesting for the stations with similar site characteristics, i.e. rural Asian and rural European stations, remote Asian and remote European stations.
3. Results and discussion

3.1 Trend Assessment

There are statistically significant upward or downward trends found for about a half of considered stations. Numbers of both types of trend behavior are approximately equally. The sites with decreasing/increasing/absence trends for individual compounds as parts of total number of stations are presented at Figure 2 (a, b) for entire observational data and seasonal subsets with green/red/grey colors correspondently.

![Figure 2](image_url)

**Figure 2.** Parts of remote (a) and rural (b) stations with calculated decreasing (green), increasing (red) or insignificant (grey) trend for measured compounds to total number of stations.
Figure 3 (a, b) presents an example of illustrated results of trend assessment for sulfate been put on the map. Similar pictures were prepared and analyzed for all selected species. The description of results is given below.

![Spatial distribution of SO4.2m trends at EANET monitoring stations in North-East Asia](image)

(a) Figure 3. Spatial distribution of entire observational record (a) and seasonal (b) trends of sulfate. Empty circles denote not significant trends. Color and its intensity represent direction and absolute value of the trend for the station: red tones indicate increasing trends, green indicate decreasing trends.
Aerosol sulfate trends illustrated at Figure 3 (a, b) indicate the increase of concentrations at central part of the region (Primorskaya, Kanghwa, Oki, Rishiri) while decreasing trends were evaluated for western part of the region as well as for eastern except Ogasawara. The same is for seasonal trends.

Filterpack measurements for sulfur dioxide were available only for four stations (three Russian and Mongolian). For all the stations there are increasing trends but seasonal trends are different. For example there are no statistically significant trends for autumn season. Highest increasing trend shows Mondy during winter season (increase about 0.5 ppb per year).

Whole situation for nitric acid is not so clear. For entire observational record trends one can see significant increase only for Primorskaya station. Other trends are decreasing or increasing but values of increase per year are less than one percent compared to mean value for whole time series at this station. Same nonhomogenity in trend behavior is true for seasonal trends. Only for summer we can see strongly increasing trend for Imsil.

Entire observational records of HCl show increasing trends for western part of region of interest. Same trends are true for seasonal trends. Also there is increasing trend at Japanese station Ogasawara for spring.

Entire observational records of ammonia show decreasing trends for central part of the region, whereas Cheju and Hedo show increasing trends. Spatial distribution for autumn and spring and summer trends is similar, however for winter the only increasing trends exist for western part of the region (Listvyanka, Mondy, Terelj).

At majority of stations we see increasing trends of NO$_3^-$, especially in central part of the region. Exceptions are Mondy, Listvyanka, Ochiishi that show decreasing trends. Seasonal trends are similar.

For chloride we can see increasing trends along the line from north-west to south-east (Terelj, three Korean stations, Oki, Ogasawara). Other stations show no significant trend or slightly decreasing. Seasonal trends are similar to entire observational record.

Situation for ammonium is similar to chloride in central part and Ogasawara there are increasing trends, but there are more stations showing decreasing trends. Seasonal trends are similar to entire observational record.

For Na$^+$ central part of the region and Ogasawara show increasing trends and northern part of stations show decreasing trends. Seasonal trends are similar to entire observational record trends.

K$^+$ entire observational record majority of stations show decreasing trends and only Ogasawara shows strongly increasing trend. Seasonal behavior is very similar except increasing trend at Primorskaya in spring and increasing trend at Oki in winter.

Group of three stations (Primorskaya, Oki, Ogasawara) show increasing trends for entire observational period as well as for seasonal trends for Mg$^{2+}$. Other stations show statistically significant decreasing trends or not significant trends.

Whole situation for Ca$^{2+}$ is same as for Mg$^{2+}$ except Kanghwa that also shows increasing trend.

For majority of pairs station-species seasonal trends and entire observational record trends are similar. However there is a number of cases where there is no any statistically significant trend for entire observational record while there are two significant opposite trends for different seasons. An example of such situation is ammonia trends at remote Russian continental station Mondy. At Mondy there are statistically significant decreasing of ammonia concentrations in spring and summer significant increasing in winter and no significant trend for entire record.

Necessary to point out that Ogasawara shows strongly increasing trends for many species though it is very remote area. The reasons of such behavior should be considered.

All results including spatial distributions of trends and tables with trend slopes values are available under request to correspondent author.
3.2 Comparison of the EANET and EMEP monitoring data (aerosols and gaseous species)

For EANET and EMEP stations results of comparison of mean concentrations are shown in Figure 4 (a, b) and Figure 5 (a, b). Green cells denote that mean concentration at EANET station is statistically significant lower than at EMEP station; red color – higher; white – no significant difference. Brief description of the results of comparison is given below.

![Comparison of mean NITRIC ACID + NITRATE, [μg/m³], for EANET and EMEP stations](image)

![Comparison of mean NITRATE, [μg/m³], for EANET and EMEP stations](image)

Figure 4. Results of comparison of mean concentrations of Nitric acid+nitrate (a) and nitrate (b) at EANET and EMEP stations. Green cells denote the mean concentration at EANET station is statistically significant lower than at EMEP station; red – higher; white – no significant difference.
Remote EMEP stations have lower concentrations of the sum of nitric acid and nitrate comparing to EANET remote and rural stations. However, their concentrations at almost all rural EMEP stations considered here are higher than at rural EANET stations. The concentrations of nitrate are available only at three rural EMEP stations. Concentrations of nitrate at EANET stations are lower comparing to EMEP.

At the remote EANET stations average ammonium concentrations have higher values compared to remote EMEP stations. And its concentrations at remote Cheju station were even higher than those at rural EMEP stations. Rural EANET stations have lower concentrations except for station Imsil.

**Figure 5.** Results of comparison of mean concentrations of sulfate (a) and sulfur dioxide (b) at EANET and EMEP stations. Green cells denote mean concentration at EANET station is statistically significant lower than at EMEP station; red – higher; white – no significant difference.
The concentrations of sulfate were higher at almost all EANET stations compared to EMEP stations, except north-western continental EANET sites Mondy, Terelj and Listvyanka.

For sulfur dioxide rural continental station Listvyanka have higher concentrations than remote and rural EMEP stations as well as its concentrations at remote EANET stations are higher than at correspondent remote EMEP stations.

To summarize the evaluation results above we can make four groups of stations according to the site classification (remote or rural) and distance far from the sea. After that we compared the result outlines of correspondent stations from both EANET and EMEP monitoring networks. Groups of stations and results of comparison are presented in Table 1.

### Table 1. Description of comparison results between EANET and EMEP stations of each of four (remote-seaside, remote-continental, rural-seaside, rural-continental) groups.

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<tr>
<th></th>
<th>Seaside</th>
<th>Continental</th>
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<tr>
<td>Remote</td>
<td>EANET stations: Oki, Sado, Cheju; EMEP stations: Bredkalen, Greenland.</td>
<td>EANET stations: Mondy, Terelj EMEP stations: Oulanka</td>
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<td></td>
<td>Concentrations at EANET stations are higher compared to EMEP stations for all the species.</td>
<td>Concentrations at EANET stations are higher compared to EMEP stations except for sulfate (both EMEP stations are higher).</td>
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<tr>
<td>Rural</td>
<td>EANET stations: Imsil, Primorskaya EMEP stations: Leba, Zingst</td>
<td>EANET stations: Listvyanka EMEP stations: Svratouch, Sniezka</td>
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<td></td>
<td>Results are not so homogeneous For nitric acid+nitrate and nitrate Imsil has higher concentrations than Leba but lower than Zingst. Concentrations at Primorskaya for almost all cases for nitric compounds are lower than at EMEP stations. However, concentrations of sulfur compounds at Primorskaya and Imsil are higher than at EMEP stations Leba and Zingst.</td>
<td>For almost all the species concentrations at EANET station Listvyanka are lower compared to EMEP except for sulfur dioxide.</td>
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To make the comparison of trends between stations is not so simple and easy due to presence of uncertainty in trend assessing itself. To assess this trend uncertainty we used confidence intervals (CI) in our study. Therefore to compare two trends the relevant CIs have to be taken into account. The conclusion on difference of the trends could be done only if correspondent CIs are not intercepted. But if CIs do intercept we cannot say anything about trend equality. Consequently, to compare long term changes at stations of the monitoring networks we compared the existence and order of statistically significant trends. Methods to assess trends at EMEP stations were the same used for trends assessment performed for EANET and described above. Based on available data sets we compared estimated trends at EMEP and EANET stations. The example of the results obtained is presented in Figure 6.

Trends at European stations is also not homogeneous similar to EANET stations. There are statistically significant increasing and decreasing trends for European stations. The orders of trend magnitude are the same for compared Asian and European stations. Moreover, when we compare the relative steepness of the trend slopes (i.e. slopes divided by the mean values) these values at Asian and European stations became even closer. There is no principal difference between trends estimated for entire observational data sets and seasonal subsets.
Figure 6. Comparison of slopes of trends at EANET and EMEP stations with correspondence confidence intervals for entire observational records (a); comparison of slope to mean ratios at EANET and EMEP stations with correspondence confidence intervals for entire observational records (b).
4. Conclusion

Multi-year general and seasonal longterm changes of main acidifying components were investigated in our study based on regular measurements of their atmospheric gaseous and aerosol concentrations at EANET stations over North-Eastern Asia. There are more than a half of all stations over the whole territory of interest which trends are found to be statistically significant. However, the ratio of increasing and decreasing trends is approximately equal. Trend analysis of separated seasonal datasets provided more detailed description of long term change processes at areas of stations. For some places the completely opposite direction trends had been recognized for different seasons while trend for common data of entire period was estimated to be not significant. Spatial peculiarities of trends are certainly different for the selected species but we recognized to be possible to distinguish two directions within region along each of those the trends at stations are mostly similar: from North-West to South-East, and from South-West to North-East.

Levels of pollutant concentrations at EANET stations in Asia are not homogeneous as well as ones at European stations. A comparison of pollution levels might allow us to conclude that mean air concentrations of main compounds at EANET stations are higher than at EMEP stations for majority of cases. However, a number of opposite cases was also found.

The statistically significant increasing and decreasing trends were found in monitoring data on airborne acidifying compounds at both North-East Asian and European stations. Absolute values of the trend linear slope vary among the stations and depend on pollution species at each region. Taking into account the confidence intervals for trend estimation the valid conclusion cannot be done whether absolute values of either increasing or decreasing trends are higher at EANET stations comparing to EMEP region or not. However, the orders of magnitude of trends are similar for whole sets of observational records as well as for seasonal subsets.

References

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