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Overview of the tropospheric ozone problem: formation, measurements, trends, and impacts (Hungarian specialties)

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Abstract—Ground-level or tropospheric ozone (O_3) is an oxidant air pollutant that has harmful effect on human health and vegetation, however, it is a short-lived greenhouse gas. Ozone is a secondary pollutant; which means that it is not directly emitted in the ambient air, but also produced from the photochemical oxidation of non-methane volatile organic compounds (NMVOCs), methane (CH_4), or carbon monoxide (CO) in the presence of nitrogen oxides (NO_x). It is destroyed both photochemically and through deposition to the surface. Summarizing the chemistry of ozone is complex and non-linear. Background concentrations of ground-level ozone in Europe do not show a significant downward trend, but in Hungary essential reduction (–0.28 μg/m^3) was observed at K-puszta station in the last decades. In the monthly distribution the amplitude decrease with increase in altitude, at K-puszta 45.1 μg/m^3, while at Nyírjes 36.6 μg/m^3 amplitudes were observed. Based on our data we found that the ozone gradient is about +1.4 μg/m^3/m. Breathing ozone can result in a number of negative health effects that are observed in relevant segments of the population. Ozone also is known as the air pollutant most damaging to agricultural crops and other plants. This article gives a general overview of the ozone problem focusing on the Hungarian specialties.

Key-words: air quality, ground level ozone measurements, AOT40, trends, vegetation

1. Introduction

In recent decades, ozone has been received increasing attention in the analysis of the regional and local air quality. In addition to being very important for sustaining life near the Earth's surface by absorbing hazardous UV radiation within the stratosphere, ozone is one of the most important greenhouse gases (*Paoletti* and *Cudlin,* 2012).

In the troposphere it is a strong oxidant air pollutant affecting human health and natural ecosystems, and reducing crop yields (*Wilkinson et al.,* 2012). Its concentration has doubled between the end of the 19th century and the 1980's. The annual average in the Northern Hemisphere has risen to 60–90 $\mu g/m^3$, and recently it increased with a further 10 $\mu g/m^3$ (*Wilson et al.,* 2012). In spite of the international agreements aimed to decrease the precursor gas emission, its concentration has been increasing (*Derwent et al.,* 2003, *Dentener et al.,* 2005). The reasons of the increase of ground level ozone concentration have not yet been understood well scientifically, but likely the sectors such as international shipping and air transport could be responsible for it, because the emissions of precursor gases from these activities are not regulated strictly enough (*Dentener et al.,* 2005). Since the sources of ozone are not confined to a smaller area but can be found worldwide, the problems related to ozone pollution has to be managed globally. Ground-level ozone episodes, when O_3 concentrations may reach at 400 $\mu g/m^3$ or more, occur in polluted regions under hot and sunny conditions. This has been ascribed to photochemical processes due to the occurrence on a large scale of favorable meteorological circumstances (*Guicherit* and *Van Dop,* 1977). The long-range transport of tropospheric ozone and its precursors have important impact on O_3 concentrations at regional and local scales. In the low to midlatitudes, O_3 transport from the polluted source regions like North/South America, Europe, and Asia generally accounts for more than 50% of ozone even in remote locations (*Sudo* and *Akimoto,* 2007).

The Council Directive on air pollution by ozone (92/72/EEC) defines several threshold levels, and it establishes a harmonized procedure for monitoring and exchanging data. It also arranges to provide the public with information when warning and information threshold levels are exceeded.

- Health protection threshold: days with an 8-hour average ozone concentration of more than 120 $\mu g/m^3$.
- Population information threshold: the hourly average ozone concentrations exceed 180 $\mu g/m^3$.
- Population warning threshold: the hourly average ozone concentrations exceed 360 $\mu g/m^3$.

Air pollution is a process that introduces diverse pollutants into the atmosphere that cause harm to humans, other living organisms, and the natural

environment (*Brauer et al.,* 2012; *Kim et al.,* 2013). The effects of O_3 found that exposure to ambient O_3 levels is linked to such respiratory ailments as asthma, inflammation, and premature aging of the lung, and to such chronic respiratory illnesses as emphysema and chronic bronchitis (*Delfino et al.,* 1998). More than two million deaths are estimated to occur globally each year as a direct consequence of air pollution through damage to the lungs and the respiratory system (*Shah et al.,* 2013). Among these deaths, around 2.1 and 0.47 million are caused by fine particulate matter (PM) and ozone, respectively (*Chuang et al.,* 2011; *Shah et al.,* 2013).

Besides positive CO_2 and nitrogen fertilization, many studies have show that ozone and its precursors are efficiently transported in the regional scale and consequently ozone tends to present relatively high background levels in rural areas. Over 90% of vegetation damage may be result of tropospheric ozone alone (*Adams et al.*, 1986). Previously, it was thought that tropospheric ozone is an urban problem, elevated O_3 concentrations are now recognized as extending far beyond city limits. Elevated concentrations in rural regions significantly affect crop yields, forest productivity, and natural ecosystems. Evaluations of the national economic impact of ozone on crop yield have indicated values of the order of US$ 2-4 billion in the United States and of 4 billion EUR in Europe (*Murphy et al.,* 1999, *Holland et al.*, 2002).

Ozone in the ground level is also expected to contribute to the devastation of building and material. In developed countries, where the control of emissions of air pollutants are relatively efficient, and the emission projections for the precursor gases indicate continuous decrease (*Kelly et al.,* 2010), the O_3 concentration will likely decline in the next decades. Although the climate change will complicate the picture the rate of decline will be slowed down. Due to the climate change heat waves may occur more often and could cause extremely high ozone concentrations for a short time. This fact could also demonstrate that while O_3 as a greenhouse gas affects the climate, the climate change will result weather conditions which may lead to the elevation of O_3 concentration in the ground level. It also shows that the climate change is one of the most complex problems, and this also proves the importance the international cooperation to solve the issue of air quality. Currently, in Europe the revised Gothenburg protocol specifies emission reduction commitments for precursors. In case of Hungary the emission reduction of volatile organic compounds (VOCs) was committed by 30 percent as NO_x by 42 percent, both of them are precursor gases of ozone.

1.1. Ozone formation and air quality

Ozone is found in two different areas of the atmosphere – the stratosphere and the troposphere. In the stratosphere, ozone provides a protective shield by filtering out the dangerous ultraviolet radiation from the sun. Here the ozone

molecules are formed by the photo-dissociation of molecular oxygen, and the atomic oxygen reacts with molecular oxygen to produce ozone.

Tropospheric ozone is not emitted directly in to the atmosphere, so it is described as a secondary air pollutant. It is an important atmospheric oxidant, smog component, and a short-lived greenhouse gas. In the troposphere, it is formed by photochemical reactions of nitrogen oxides with volatile organic compounds, methane, and/or carbon monoxide in the presence of sunlight. NO_x is primarily a product of fossil fuel combustion (63%), but secondarily it is a result of biomass burning (14%) (*IPCC,* 2001). Natural vegetation is a source of VOCs, which decompose into peroxy radicals, which react with NO to produce NO_2. In urban regions with high concentrations of NO_x, ozone production is generally VOC-limited, whereas in suburban or rural regions with low NO_x levels, ozone production is NO_x-limited. The different spatial distributions of NO_x and VOC production, as well as NO destruction of ozone, often result in the largest ozone concentrations downwind of urban centers, rather than in urban areas themselves (*Gregg et al.,* 2003). Transport of the chemical mixtures eventually results photodissociation and O_3 transformation at sites quite distant from original source of the precursor emissions. Increasing rate emissions of its precursors have caused ozone concentrations to experience a strong increase in highly populated continental regions during the last century (*Marenco et al.*, 1994). Tropospheric ozone concentrations can also be influenced by UV radiation. A rise in UV radiation intensity is expected to decrease tropospheric ozone in a clean environment, while increased UV in regions with precursors (NO_x, CO, CH_4) rich atmospheres will lead to increased tropospheric ozone concentrations. Mainly in summer months, when conditions of O_3 formation improve (high temperature, radiation, wind stagnation), O_3 levels typically rise even in remote background areas. Ozone is, therefore, no longer just an issue for air quality, but a complex environmental problem.

1.2. Ozone measurements and trends in Europe and Hungary

The analysis of European rural background ozone trends between 1996–2005 had carried out by *Wilson et al.* (2012). They processed data available from the EMEP and GAW monitoring stations and concluded that, on the European scale only slight increase in ozone levels (0.32–0.04 $\mu g/m^3$ per year) with a total range of 2.56 to 2.1 $\mu g/m^3$ per year can be determined. The greatest reduction is observed at K-puszta, Hungary (–4.11% per year). The first trend analysis using the tropospheric ozone measurements from K-puszta was published by *Haszpra et al.* (2003). In this paper, 0.64 $\mu g/m^3$ per year increasing ozone trend was determined for the time period of 1990–2002.

In Hungary, the Hungarian Meteorological Service is responsible for the rural ground-level ozone measurements. The institute maintains three background monitoring stations, where tropospheric ozone measurements are

carried out beside the observation of other pollutants. Since the stations are located in different geographical environments from plains to mountains, this effect is reflected in the measured data. *Fig. 1* shows the positions of the monitoring stations in Hungary.

Farkasfa background air pollution monitoring station is located in the western part of Hungary (46°54'37" N, 16°18'34" E, 312 m asl), at the area of the Őrség National Park. The station is surrounded by forest and no essential local source can be found nearby. The tropospheric ozone measurements started at this station in 1996. For many reasons, the ozone measurements were interrupted between 2005–2006, but in May of 2006, the operation of the station restarted.

K-puszta is the regional background air pollution monitoring station located in the central part of Hungary, on the Hungarian Great Plain (46°58' N, 19°33' E, 125 m asl). The station is located in a big forest clearing. In the wider region, agricultural fields, forest patches, pastures, and open bushy regions can be found. The prevailing wind blows from the west-to-north sector. The nearest town (Kecskemét, approximately 112 thousand inhabitants) is about 15 km to the southwest. The tropospheric ozone measurements started at this station in 1990. K-puszta monitoring station belongs to the European air quality monitoring (e.g., EMEP) and the Global Atmosphere Watch (GAW) networks. The station was also involved in the Tropospheric Ozone Research, a Sub-Project of EUROTRAC project (*Haszpra et al.,* 1997).

Nyírjes background air pollution monitoring station is located in the Mátra Mountains, in the northeast part of Hungary (47°52’ N, 19°57' E, 702 m asl). The tropospheric ozone measurements started at this station in 1996.

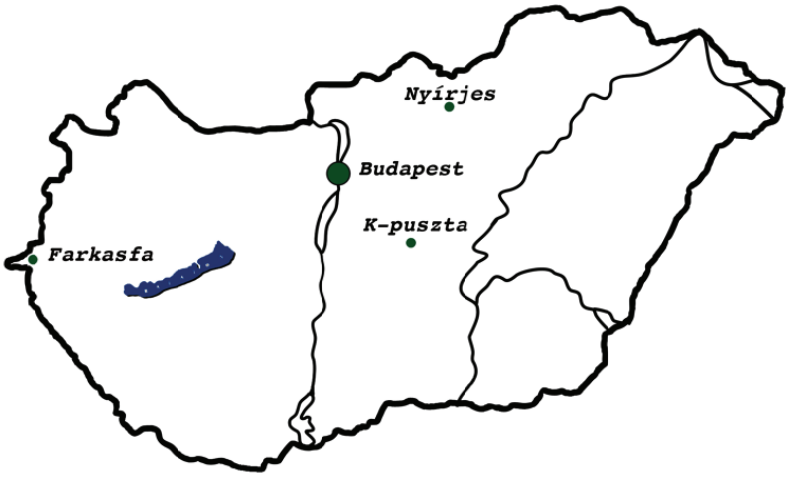

Fig. 1. Location of the ground-level background ozone monitoring stations in Hungary

2. Results

2.1. Annual trend in background stations

This part concerns the background stations for the annual trend analysis over the 1996–2014 period. Atmospheric lifetimes of ozone precursors are long enough to allow them to be transported on long distance, but the range of the impact depends on meteorological and geographical conditions. Although the stations are located in background areas, the local topography and surroundings are different. These circumstances undoubtedly may influence the ozone concentration. *Fig. 2* shows the calculated ozone trends for the Hungarian monitoring stations. We assumed that the trends are linear calculated by deseasonalizing the ozone time series. Mann-Kendall analysis of Sen-Theil slopes was used for the calculations.

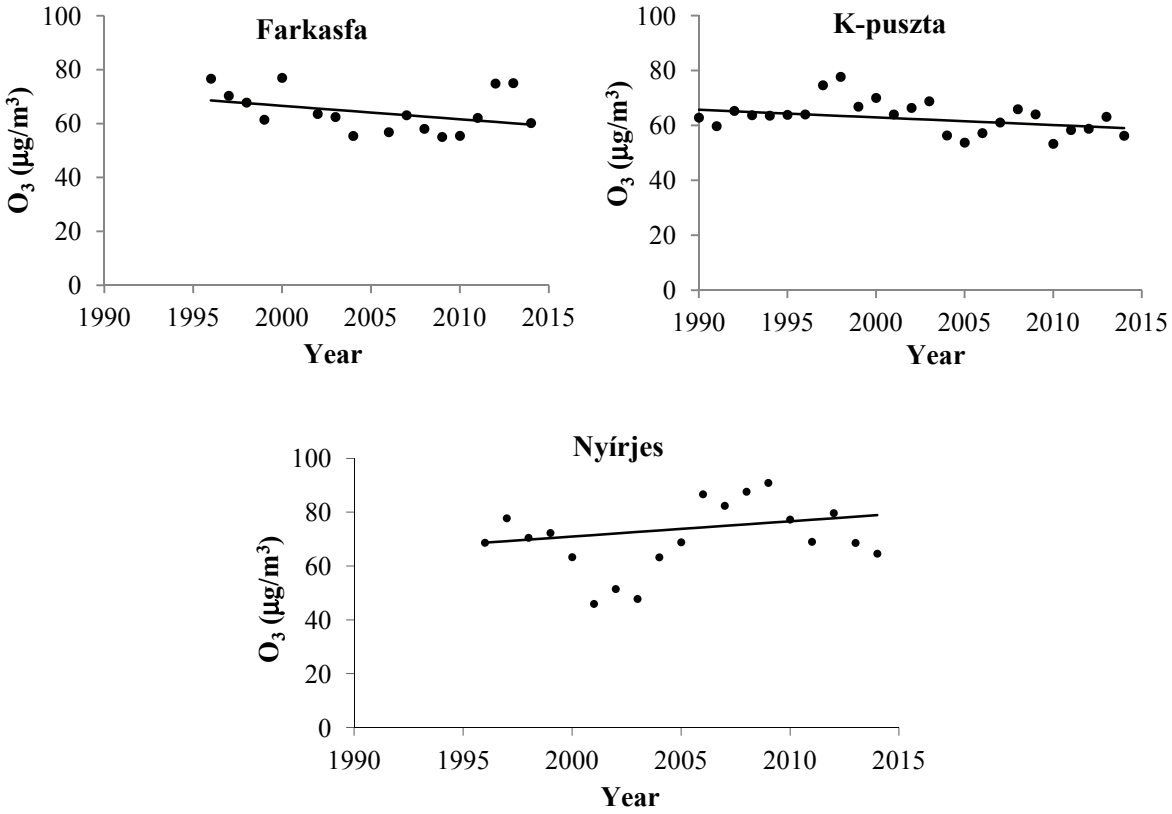

Fig. 2. Ozone trends detected at the Hungarian background air quality monitoring stations (dots = annual mean).

In case of Farkasfa and K-puszta stations, decreasing trends can be observed, while in case of Nyírjes station, the trend is increasing in average but there are intervals when the ozone concentration decreased for a few years (between 1997–2002 and 2009–2014). The trend determined for K-puszta using

our 25 years data is –0.276 μg/m^3 per year, but the trend determined by *Wilson et al.* (2012) using only 10 years (between 1996–2005) shows more intensive decreasing (–1.826 μg/m^3 per year). Similar results were found by *Sicard et al.,* (2013) over the period 2000–2010, when annual mean concentrations decreased by 0.43% per year at rural sites. Explanation of the maximum values and evolution could be in the local and annual meteorological conditions. At K-puszta, the concentration values usually higher than at Farkasfa that can be explained on one hand by the different meteorological conditions (higher temperature) and higher altitude on the other hand by the ozone plumes coming from Budapest city (*Mészáros et al.,* 2009). The decreasing trends at Farkasfa and K-puszta can be attributed to the reduction in NO_x and VOC emission within Europe. Background concentrations of ozone in Europe are influenced significantly by emissions of precursor gases outside the continent (*Guerreiro et al.,* 2014). Changes of the annual mean of ozone concentrations could have been caused by the precursor gas emission, the effect of the long-range transport of ozone and pre-gases, and the meteorological situation (heat waves, rainfall). *Fig. 3* shows the abatement in NO_x and VOC levels in Hungary that could have caused the reductions in episodic peak ozone levels.

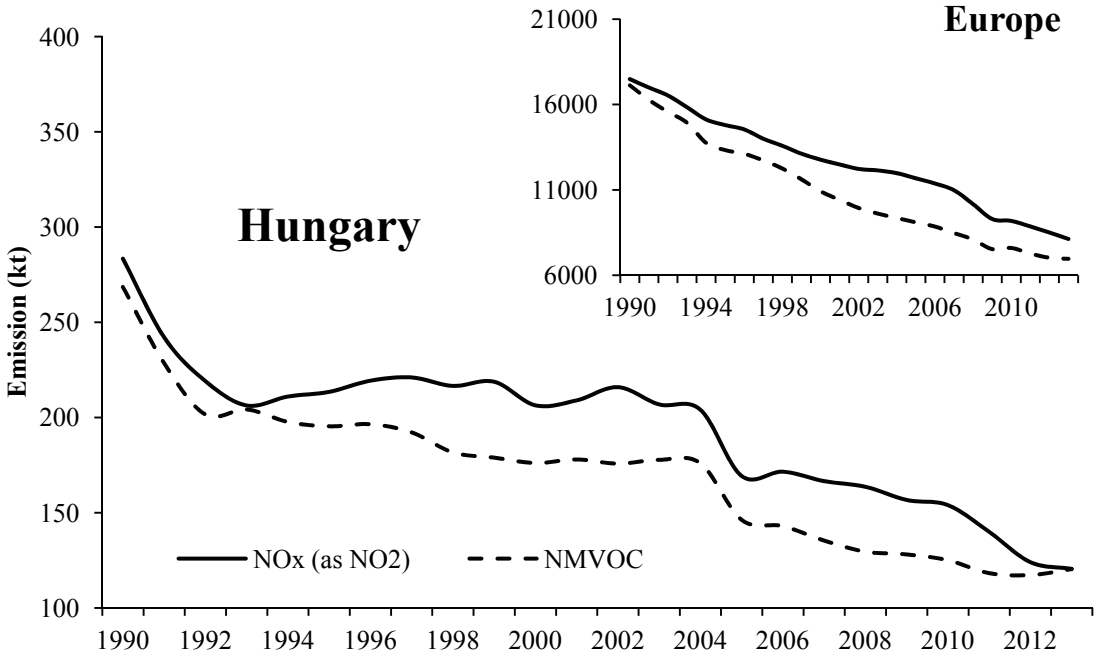

Fig. 3. Anthropogenic NO_x (as NO_2) and NMVOC emissions 1990-2013 for Hungary and Europe (Source: www.ceip.at).

According to *Sicard et al*. (2009), between 1995 and 2003, a rate of –0.48% per year for stations below 1000 m asl and +1.75% per year for stations above 1000 m asl were observed. They mentioned that the possible explanations could be the following:

1.) If ozone is produced in lower field from exhaust and industrial sources, it goes up with the favour of the temperature inversion phenomenon. At high altitude, ozone stagnates to form a reservoir layer.

2.) Altitude sites are not influenced by the ozone destruction by nitrogen oxides. Indeed, the NO concentration, emitted mainly by the road transport, are weak in the higher altitude. In addition, there are biogenic VOC emissions, emitted by the vegetation, which can increase the ozone production.

3.) Approximately 10% of tropospheric ozone is estimated to be of stratospheric origin.

It is also known that background ozone level increases with the height in the lower troposphere. Based on our data (average ozone concentration from 1996 to 2014), a change in the ozone vertical gradient is clearly visible (see *Fig. 4*). The ozone gradient is about +1.4 μg/m^3/m. This spatial distribution shows an interesting pattern, because the most urban-influenced site (K-puszta) has the lowest concentration, while either the distance or the elevation is not more efficient contribution to reach this high ozone concentration. However, this problem is quite complex, further investigation is needed to explain these findings.

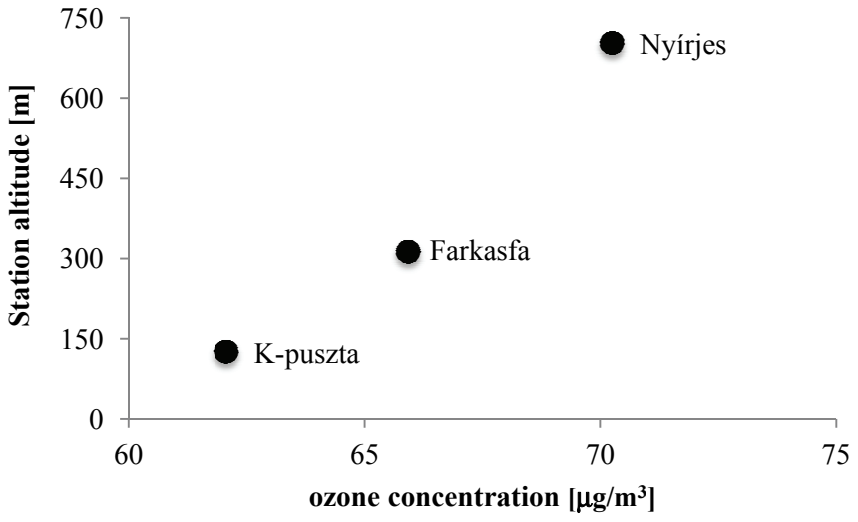

Fig. 4. The total average ozone concentration depending on the station altitude.

2.2. Seasonal trend analysis

The monthly means of ozone concentration were also determined for the three stations. *Fig. 5* presents the results of this investigation. The three diagrams

reflect that the ozone has different yearly variation on the sites. The biggest amplitude can be observed at K-puszta (45.1 µg/m^3), while the lowest at Nyírjes (36.6 µg/m^3). This result is reflects the fact that Nyírjes is a mountain station where the amplitude of the monthly and daily ozone concentrations are much lower than at the plain stations (*Chevalier et al.,* 2007).

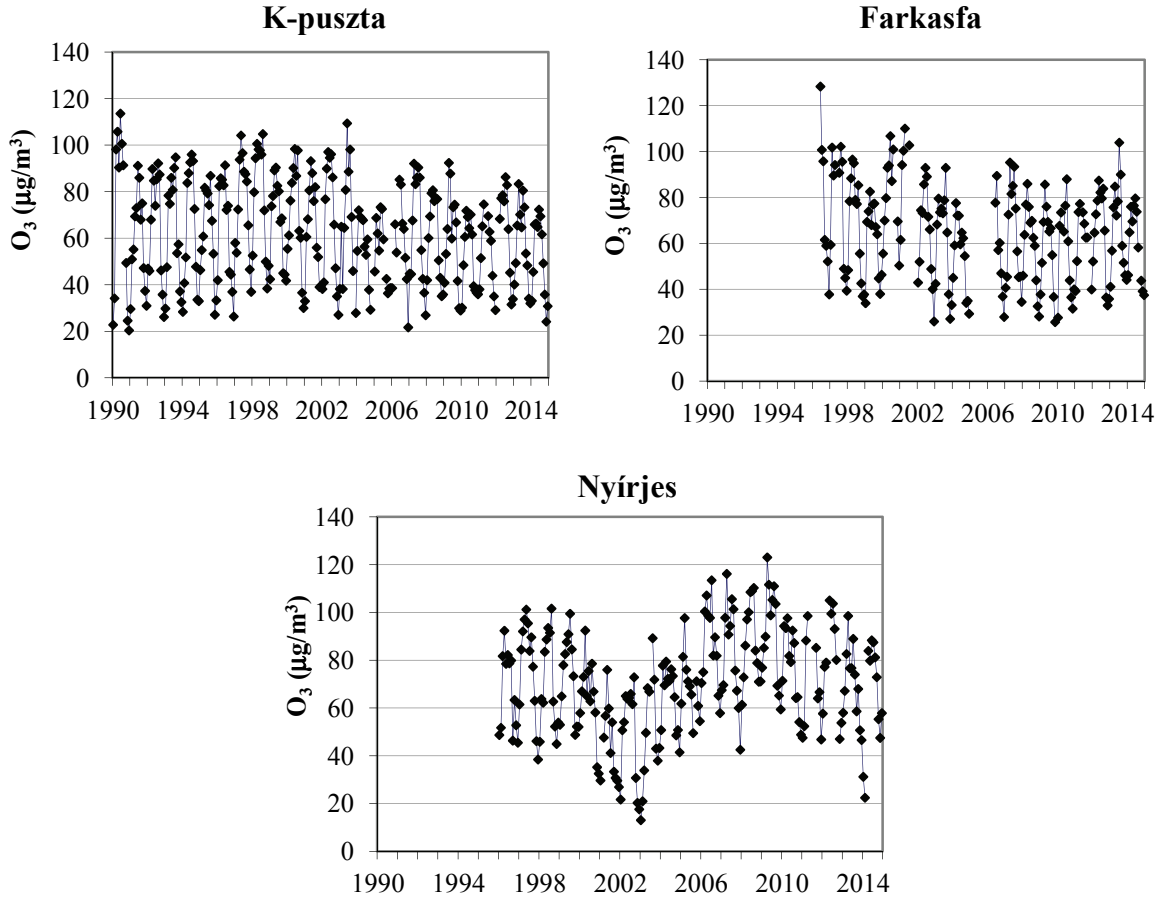

Fig. 5. Temporal variation of observed monthly mean ozone concentrations detected at the Hungarian background air quality monitoring stations (dots = monthly mean).

The trends for the 5th, 50th, and 95th percentiles of ozone concentrations show a decrease for Farkasfa and K-puszta while an increase for Nyírjes (*Fig. 6*). In case of Farkasfa and K-puszta, the 95th percentile decreasing trend is much larger and the 5th percentile decreasing trend is much lower than the median trend. It means that the decreasing trends of the yearly mean values are basically caused by the decreasing maximum concentration values, while the minimum values show a moderate rising.

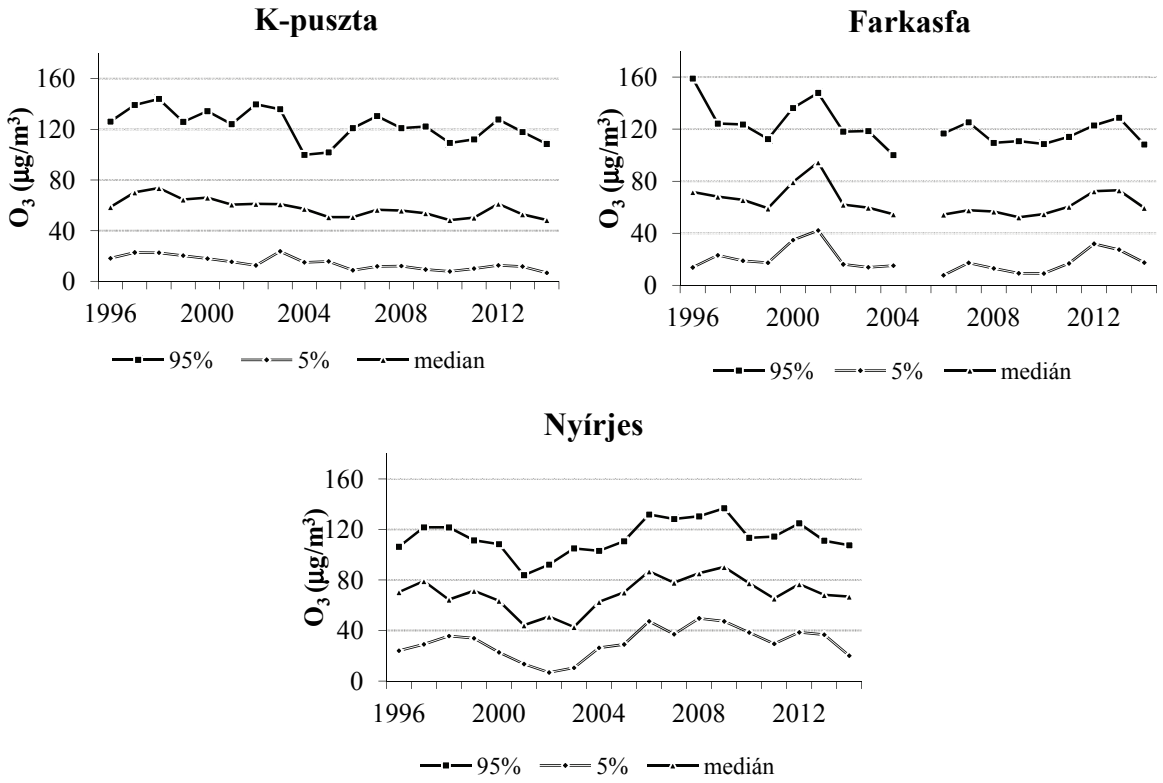

Fig. 6. Temporal variation of the annual 95th, 50th, and 5th percentiles of ozone concentrations detected at the Hungarian background air quality monitoring stations.

In case of Nyírjes, in all three percentiles (95th, 50th, 5th) a rising trend was found (see *Table 1*). The mean ozone concentration is known to strongly increase with altitude in the troposphere mainly in the first 1000 m. The comparison of the results of the three Hungarian stations also shows that the minimum ozone concentration is rising with the altitude. This can be explicable with that the ozone content is eroded near the surface by deposition and titration that dominate in the boundary layer at the yearly time-scale.

Table 1. Mean trends by stations for the 5th, 50th, and 95th percentiles of ozone concentrations

	Trend (µg/m^3/yr)		
Station	**5th percentile**	**median**	**95th percentile**
K-puszta	–0.75	–0.97	–1.24
Farkasfa	–0.20	–0.58	–1.23
Nyírjes	+0.76	0.67	+0.67

Sicard et al. (2013) found that background ozone concentration decreased of –1.1% per year with annual averages and –0.9% per year with median values. They observed that the number of concentration values above 65 μg/m^3 increased significantly (1% per year), and the 25th percentiles decreased of –0.4% per year over the 2000–2008 period. Based on these and our results the background ozone level seems to increase. However the maximum ozone concentration shows decreasing trend, there are several days when the values reach a threshold. The exceedance days of air quality threshold values for ozone at the Hungarian background air quality monitoring stations decreased in the last 25 years in connection with the emission reduction of precursor gases. The number of days when the O_3 concentration exceeds the previously defined threshold values for ozone was also determined using the Hungarian monitoring data (*Fig. 7*). At our stations, the ground-level ozone has never exceeded the population warning threshold in the examined period, but the ozone concentrations were quite often above the other threshold values. The ozone concentration exceeded the health protection threshold more than 100 times per year only at K-puszta in the time period of 1997–2002. Since K-puszta is a background station, it might be better termed "urban-affected", because of advected urban plumes from Budapest that affect concentration characteristics. Because Nyírjes and Farkasfa are beyond the reach of urban plumes or other anthropogenic effects, they can show small seasonal variations in ozone concentration.

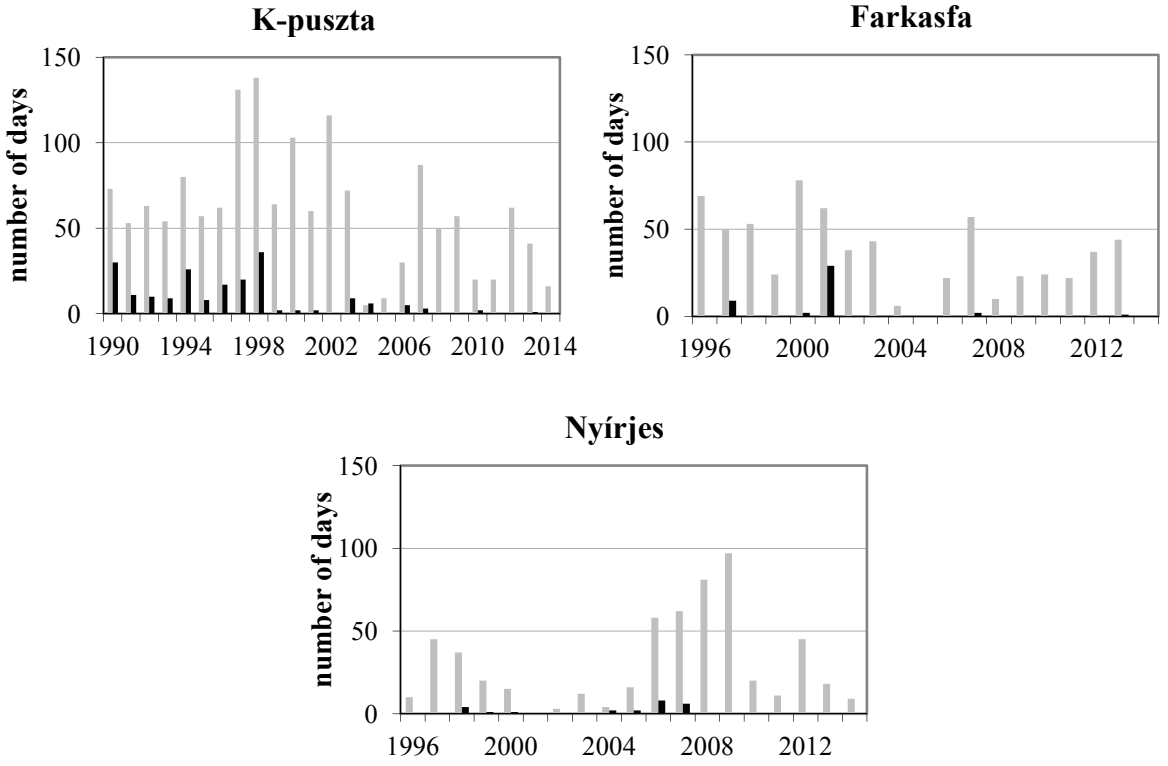

Fig. 7. Exceedance days of air quality threshold values for ozone at the Hungarian background air quality monitoring stations (gray = health protection threshold, black = population information threshold).

The frequency distribution of the hourly ozone concentrations was also calculated. For the comparability of the results, we used only the data from the time period 1996–2014 at all three stations. From many European country's data it can be concluded, that the proportion of ozone concentration range of 40–78 μg/m^3 has increased, while the proportion in the category, 80–118 μg/m^3 has not changed in the summer months. The main differences in terms of frequency distribution can be found in the winter season between the mountain and plain stations. *Fig. 8* shows the results of this examination. The data availability for the stations and for the time period of 1996–2014 was: Farkasfa 80%, K-puszta 90%, Nyírjes 86% (in case of K-puszta, the data availability was 87% for the time period of 1990–2014). The highest ground level ozone concentrations can be expected at Farkasfa in June and July, at K-puszta in June, July, and August, while at Nyírjes only in July.

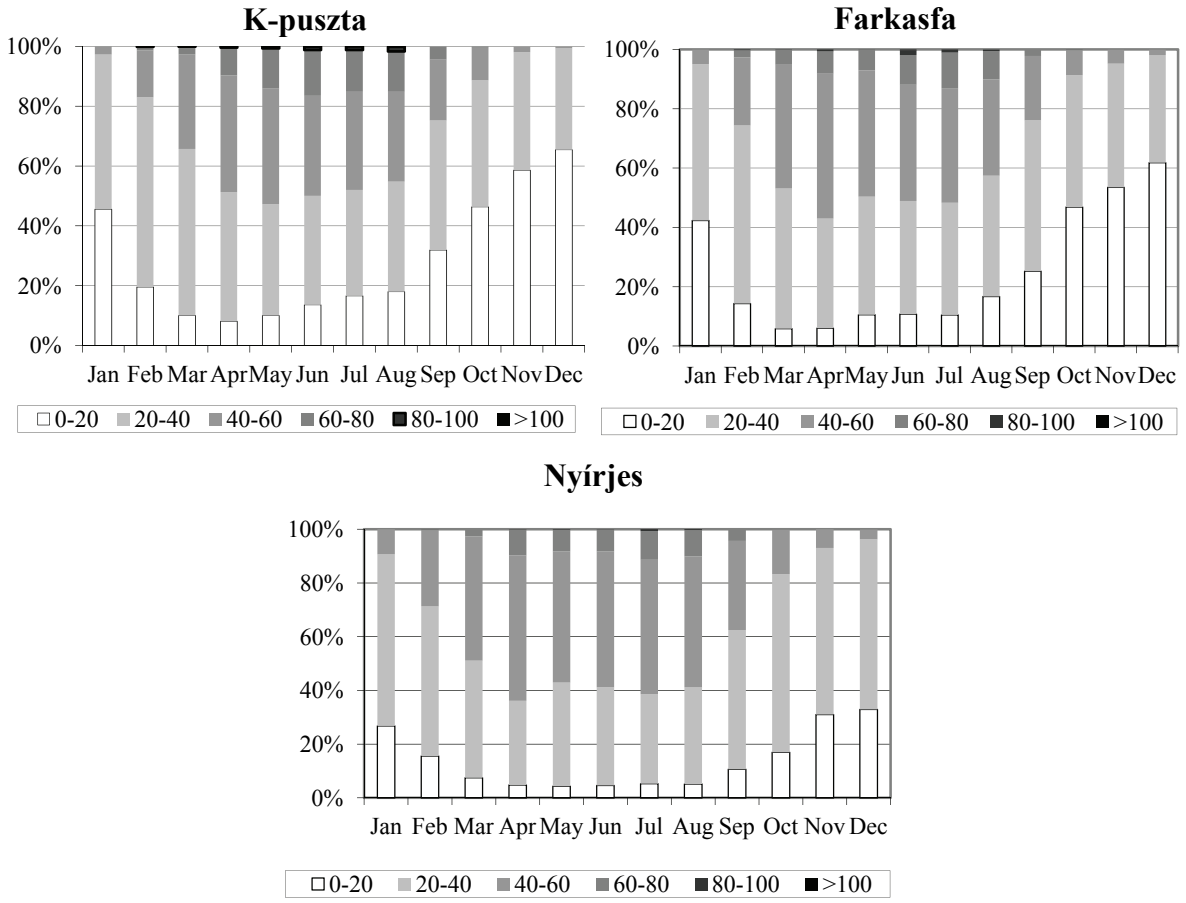

Fig. 8. Frequency distribution of ozone concentrations (hourly values) at the Hungarian background air quality monitoring stations.

2.3. AOT trends

Among the European standards, AOT40 index is generally used for the protection of the vegetation. AOT40 is defined as the sum of differences

between the hourly mean concentration and the 80 µg/m^3 threshold value for each hour when the concentration exceeds 80 µg/m^3. Then these values are summarized each day from May 1 to July 31, for the time period of 8–20 hours. In this study we found significant differences among the stations (see *Fig. 9*).

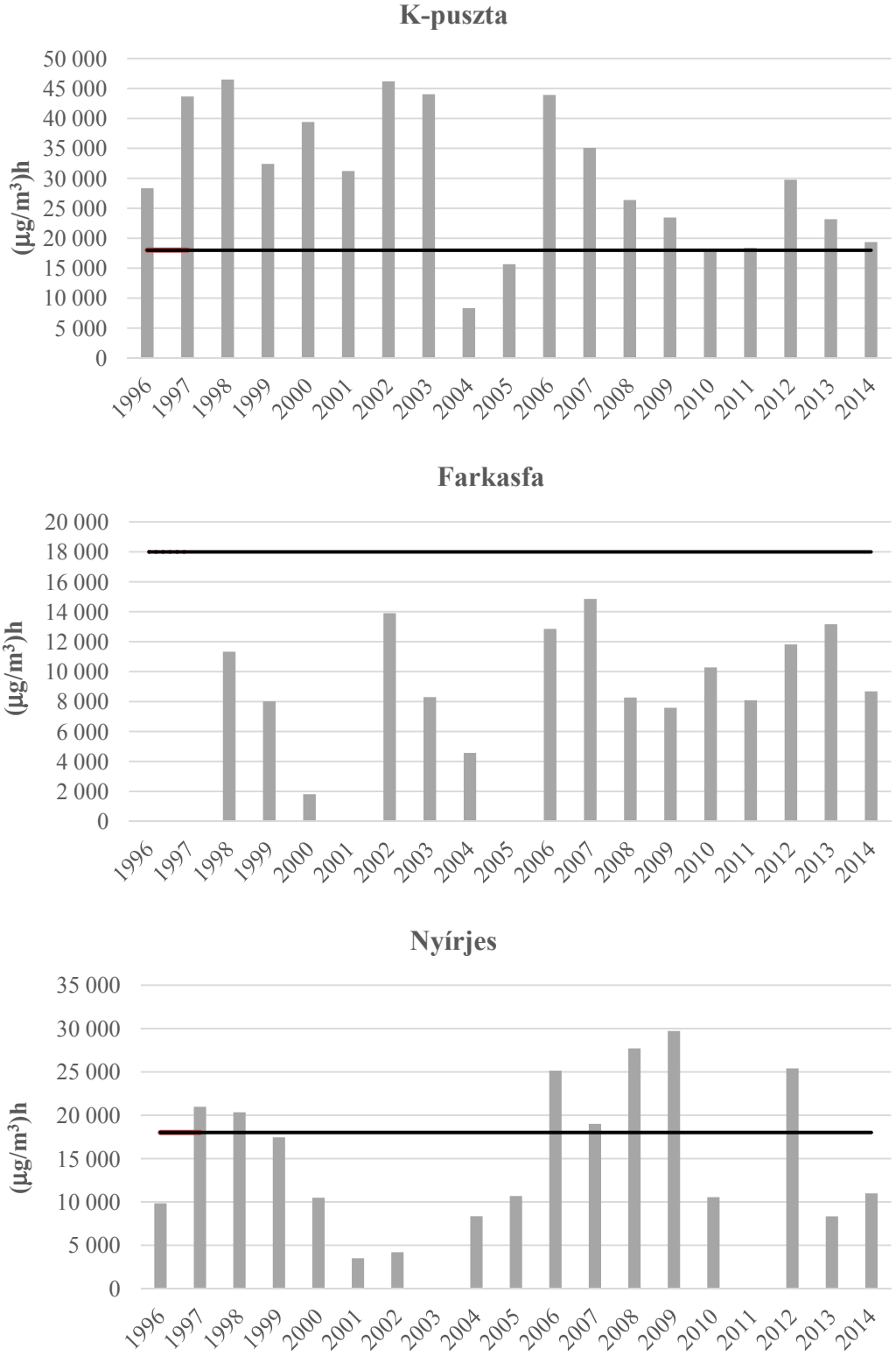

Fig. 9. AOT40 values (µg/m^3)h at the Hungarian background air quality monitoring stations (horizontal black line = threshold value).

Despite the Hungarian declining ozone concentration trends (see *Fig. 2*), AOT40 did not show any trend. In case of K-puszta, the AOT40 exceed the limit value in most cases, while at Farkasfa the calculated AOT40 has never been exceeded the limit value, and at Nyírjes the AOT40 exceeded the limit value only in some cases. Nyírjes and Farkasfa stations are located in areas devoid of local pollution sources, which can be a possible reason for the AOT40 values. In contrast, K-puszta is located in the direction of Budapest's plume, and due to its geographic position, the annual average temperature is higher than in the other stations, which favors for the higher ozone concentration, consequently for higher value of AOT40.

Between 21% and 69% of agricultural crops in the EEA-32 (European Economic Area) were exposed to O_3 levels above the EU target value for protecting vegetation (18 000 ($\mu g/m^3$)h for AOT40) from 2002 to 2010, mostly in southern and central Europe (*EEA*, 2013). Reduction of yield with increasing ozone over a 80 $\mu g/m^3$ threshold, resulting in a 10% reduction in yield for ozone levels commonly found in southern Europe (*Fuhrer et al,* 1997). In Hungary, mainly the beans have shown ozone-injury symptoms.

3. Conclusion

In this paper, recent results on ozone levels and trends at background sites located in Hungary are discussed. Studies have shown that concentrations of ozone in the Hungarian background stations are influenced by emissions of precursor gases outside the continent. The Mann-Kendall test was used to detect the trend from background ozone concentrations. In case of Farkasfa and K-puszta stations, decreased trends (–0.498 $\mu g/m^3$ and –0.277 $\mu g/m^3$) can be observed, while in case of Nyírjes station, the trend is increasing (0.567 $\mu g/m^3$) in average, but there are intervals when the ozone concentration decreased for a few years. In the monthly distribution, the ozone concentrations show different variation. The amplitude decreased due to the increase in altitude, at K-puszta 45.1 $\mu g/m^3$, while at Nyírjes 36.6 $\mu g/m^3$ amplitudes were observed. Since K-puszta is a background station, it might be better termed "urban-affected", because of advected urban plumes from Budapest that affect concentration characteristics. Nyírjes and Farkasfa are beyond the reach of urban plumes or other anthropogenic effect, they can show small seasonal variations in ozone concentration. Based on our data, we found that the ozone gradient is about +1.4 $\mu g/m^3/m$. We observed that in case of Farkasfa and K-puszta, the 95th percentile decreasing trend is much larger and the 5th percentile decreasing trend is much lower than the median trend. It means that the decreasing trends of the yearly mean values are basically caused by the decreasing maximum concentration values. According to AOT40, we found big differences between the stations, however, the data did not show any trend. As Nyírjes and Farkasfa

are located in areas devoid of local pollution sources, the values of AOT40 were low, in contrast to K-puszta, where the AOT40 were higher in every year, consequently.

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