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Comparison of 19th Century and Present Concentrations and Depositions of Ozone in Central Europe

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Abstract – Ozone, one of the most important trace gases in atmosphere was discovered by Christian Friedrich Schönbein (1799–1886), a chemistry professor at the University of Basel. The method developed by him was used from the middle of nineteenth century until the 1920's in much of the world. The measurement method is based essentially on the color-change of an indicator test paper. We obtained records for ozone measured in the Habsburg Empire using Schönbein's method for analyze the long term environmental processes. According to records kept in the Habsburg Empire, ozone was measured at more than twenty sites between 1853–1856. On the territory of the Kingdom of Hungary, ozone was measured at Szeged, Buda and Selmecbánya (Schemnitz, Banska Štiavnica) among others. Long term datasets are available from Buda (1871–1898) and Ó-Gyalla (Altdala, Hurbanovo, 1898–1905). Ozone was measured during both day- and nighttime. Additionally meteorological variables (like air temperature, relative humidity, air pressure, wind speed, cloud cover, precipitation) were also observed several times a day. The data reported in the yearbooks were collected and evaluated in this study to reconstruct the ozone dataset. Depending on concentrations and deposition velocity over different vegetated surfaces the ozone deposition can be estimated. The reliability of estimations and reconstructed ozone deposition values are also discussed. Finally ozone datasets from the 19th and 21st century and the differences in ozone concentration and deposition between rural and urban areas are compared. Ozone concentrations and deposition are found to be approximately three times higher now than in the 19th century.

ozone concentration and deposition / Schönbein's method / historical datasets

Kivonat – A jelenlegi és a XIX. századi ózonmérések összehasonlítása. Az ózon fontos légköri nyomgáz, amelyet Christian Friedrich Schönbein (1799–1886), a bázeli egyetem kémia professzora fedezett fel. Az általa kifejlesztett mérési eljárást a XIX. század közepétől az 1920-as évekig használták világszerte. Egy indikátor papír színváltozását regisztrálták tíz, illetve tizennégy fokozatú skálán. A Habsburg Birodalomban végzett feljegyzések szerint az ózon koncentrációt több mint 20 helyen mérték 1853 és 1856 között. A Magyar Királyság területén ózonmérések folytak többek között Szegeden, Budán és Selmecbányán. Hosszúidejű megfigyelési sorok vannak Budáról (1871–1898) és Ó-Gyalláról (1898–1905). Az ózont nappali és éjszakai periódusban mérték. A meteorológiai elemeket – mint a hőmérséklet, relatív nedvesség, légnyomás, szél, felhőzet, csapadék – szintén naponta többször feljegyezték. Az évkönyvekben közölt adatokat dolgoztuk fel, létrehozva egy hosszúidejű ózon-adatbázist. A koncentráció és a különböző ökoszisztémákra jellemző ülepedési sebesség ismeretében az ózon terhelést is

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meghatároztuk. Összehasonlítottuk a XIX. századi és a XXI. századi ózonméréseket. Elemeztük a városi és vidéki mérőhelyeken mért ózon koncentráció és ülepedési értékek különbségeit. Az ózon szint és az ülepedés mértéke jelenleg hozzávetőlegesen 3-szorosa a XIX. századi értékeknek.

ózon koncentráció / Schönbein módszer/ történelmi adatsorok

1 DISCOVERY OF OZONE AND ITS ENVIRONMENTAL EFFECTS

The first technique for measuring ozone was developed by Schönbein. The method was based on an indicator paper coated with starched potassium iodide. A color-change of an indicator test paper was recorded on a scale ranging from 0 to 10 or 0 to 14. The strip of paper turns brown depending on the extent of the reaction of iodine with ozone and also because of humidity. Because of the effect of humidity on the measurement, the relation between color change and the concentration of ozone is not linear (e.g., Fox 1873; Linvill et al. 1980; Kley et al. 1988; Walshaw 1990; Möller 1999; Mordecai 2001).

Ozone is beneficial in the stratosphere by absorbing UV radiation, in the same time it is a pollutant near the surface. Ozone near the surface harms vegetation and animals, it is toxic to humans; decreased agricultural crop yields and damage to man-made materials are examples among its negative effects. Ozone is a secondary pollutant meaning that it is not directly emitted into the atmosphere. Instead, ozone is produced in the atmosphere by photochemical reactions involving precursor species (such as NO, NO_2, CO, VOC) released from man-made sources (such as traffic and industry) and from natural sources (such as vegetation and lightning). Stratospheric-tropospheric exchange is another important source of tropospheric ozone (Sándor et al. 1994). The concentration of near surface ozone has characteristic seasonal and daily variations. Its seasonal variation is characterized by winter minima and summer maxima in areas that are strongly influenced by pollution sources. Secondary maxima occur in early spring due to intrusions of stratospheric air. The highest ozone concentrations are typically found in the afternoon and in the early evening as the result of photochemical activity. In remote areas, i.e., areas not strongly influenced by recent pollution, ozone has a maximum in spring and a minimum in fall. The diurnal variation of ozone is much weaker in remote areas than in polluted areas and maxima can occur at any time of the day or night (Nolle et al. 2002; Wilson et al. 2011).

Ozone is a potent greenhouse gas and is ranked third in importance after CO_2 and CH_4 (IPCC 2007). For calculating the contribution of ozone to radiative, and hence climate forcing due to anthropogenic activities, it is necessary to know the history of ozone extending from the pre-industrial era to the present. In addition, long term trends in ozone concentrations are needed to assess the long-term effects of exposure of vegetation to ozone. Currently, measurements of ozone are obtained routinely around the world by surface based measurement networks and by satellites. However, these methods have only been in use for a few decades and other techniques must be relied on to obtain semi-quantitative data for ozone.

2 CALCULATION OF OZONE CONCENTRATION AND FLUX FROM HISTORICAL MEASUREMENTS

Since shortly after the development of the Schönbein test paper method, studies have attempted to relate the color changes to ozone concentrations. Fox (1873) noted that the color change is related to humidity. This happens because a damper surface can absorb more ozone. He also found that higher temperatures lead to higher ozone. Albert-Levy (1877) realized that gases such as SO_2 lead to reduction of ozone and developed the first quantitative method for measuring ozone, based on the oxidation of arsenite. Several more recent studies have

examined the question of how to relate Schönbein numbers from the color changes to ozone concentrations (e.g., Linvill et al. 1980; Bojkov 1986; Volz–Kley 1988; Marenco et al. 1994; Pavelin et al. 1999). Wind speed and sampling time (usually 8–14 hours) can also affect the measurements. It should be mentioned that higher values of Schönbein numbers were find in case of higher wind speeds at similar relative humidity in the historical ozone dataset of Buda (not presented here). Based on data from Linvill et al. (1980) and Anfossi et al. (1991), Pavelin et al. (1999) developed a procedure to account for the dependence on relative humidity (*Figure 1.*). It is the most accepted methodology for the conversion. The estimated error is ~ 20–25%.

The ozone flux (F) can be determined over a given type of vegetation using data for ozone concentration and deposition velocity (Wesely–Hicks 2000; Lagzi et al. 2004; Mészáros et al. 2009):

$$F = v_d (C_{ref} - C_0),$$

$$C_0 = 0$$

where v_d is deposition velocity, C_{ref} is concentration at reference level, C_0 is concentration at the absorbent surface.

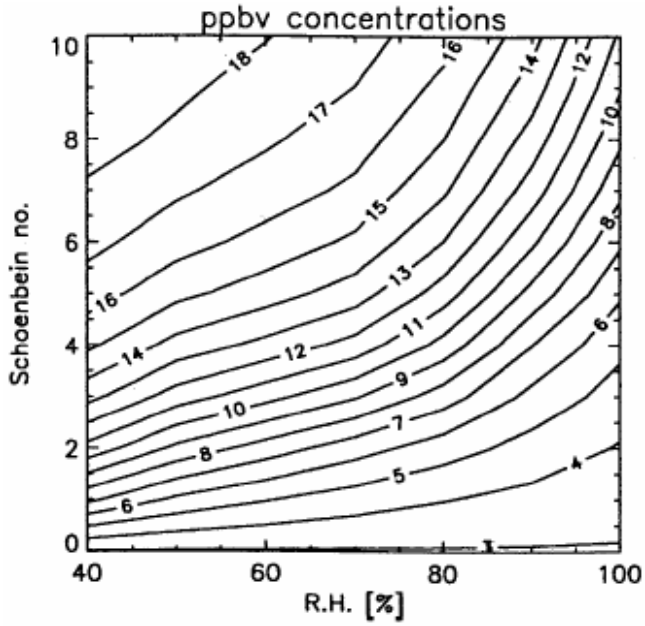

Figure 1. Humidity correction suggested by Pavelin et al. (1999) to convert Schönbein numbers to ozone concentrations in units of ppbv

Over the past few decades, numerous measurements were carried out to determine seasonal and daily variations in deposition velocity over different vegetation types (e.g. grass, deciduous, coniferous and mixed forest types). The results of these measurements could be used for parameterizing processes, or testing ozone deposition calculations using resistance models in addition to assessing ozone fluxes to vegetation (Zhang et al. 1996; Emberson et al. 2001; Lamaud et al. 2002; Lagzi et al. 2004; Büker et al. 2007).

Measurements of deposition velocity were made in the early 1990s in Hortobágy (over a grassy surface) and Nyírjes (in the Mátra Mountains over a coniferous forest) are shown in *Table 1*. Values from Nyírjes will be used for estimating ozone deposition during the vegetation growing season (from April to October) over coniferous forests. These values of deposition velocity are in good agreement (same range but little bit higher values) with other measurements found in the current literature (Padro 1996; Finkelstein et al. 2000; Zhu et al. 2008). Data shown in *Table 2* indicate that daytime deposition velocities are typically higher than nighttime

values for all the ecosystems studied. Lowest 24-hour average values were found over the grassy surface, while highest values were found over forests.

Table 1. Deposition velocity of ozone [cm/s] over a coniferous forest in the Mátra Mountains on the basis of field measurement campaigns from 1991 to 1993 (Horváth et al. 1996)

	Spring	Summer	Autumn	Winter
Daytime	0.50	1.20	0.63	0.17
Nighttime	0.04	0.34	0.03	0.04
Entire day	**0.34**	**0.86**	**0.39**	**0.12**

Table 2. Deposition velocity of ozone [cm/s] over different surface types on the basis of standard values in summer (Meyers et al. 1998; Zhang et al. 2002; Lagzi et al. 2004)

	Daytime	Nighttime	**Entire day**
Grass			**0.25** (0.2–0.35)
Agricultural	0.2 – 0.7		**0.35**
Vineyard, orchard	0.3 – 0.5	0.05 – 0.2	**0.3**
Deciduous forests	0.2 – 0.95	0.2 – 0.3	**0.5** (0.3–0.7)
Mixed forests			**0.5** (0.2–0.85)
Coniferous forests	0.7 – 0.9	0.15 – 0.35	**0.6**

3 OZONE MEASUREMENTS IN THE 19TH CENTURY

Reconstructed ozone measurements, measurement sites and measurement periods from the 19th century currently available in the literature (Pavelin et al. 1999) are shown in a contemporary geographical map (*Figure 2*). We report here reconstructed ozone measurements obtained in the Habsburg Empire from the latter half of the 19th century until the 1910s.

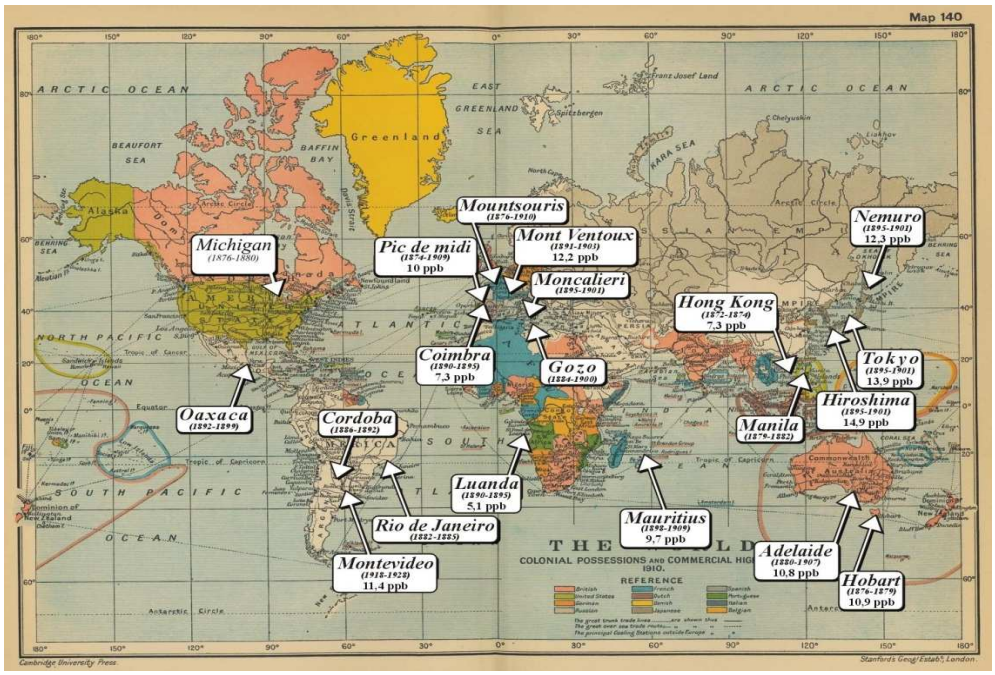

Figure 2. Ozone measurements at the end of the 19th and the beginning of the 20th century

According to the map, earliest data obtained outside the Habsburg Empire were in Hong Kong, Michigan, in France at the Pic de Midi Observatory. The measurements at each site were carried out using the Schönbein method, with different scales used, e.g., 0–10, 0–11, 0–14, 0–21. The values on the map are presented in ppbv, which has been introduced more recently. Available values were between 5 and 15 ppbv.

3.1 Ozone measurements in the Habsburg Empire between 1853–1856

It should be emphasized that the earliest datasets in the world we have are from the stations in the Habsburg Empire shown in *Figure 3*. Yearbooks between 1853–1856 of the Central Meteorological Institute of the Habsburg Empire in Vienna (Wien) contain data on ozone concentration from more than 20 stations. However, ozone data were not obtained and recorded for subsequent years, except for data obtained in Vienna.

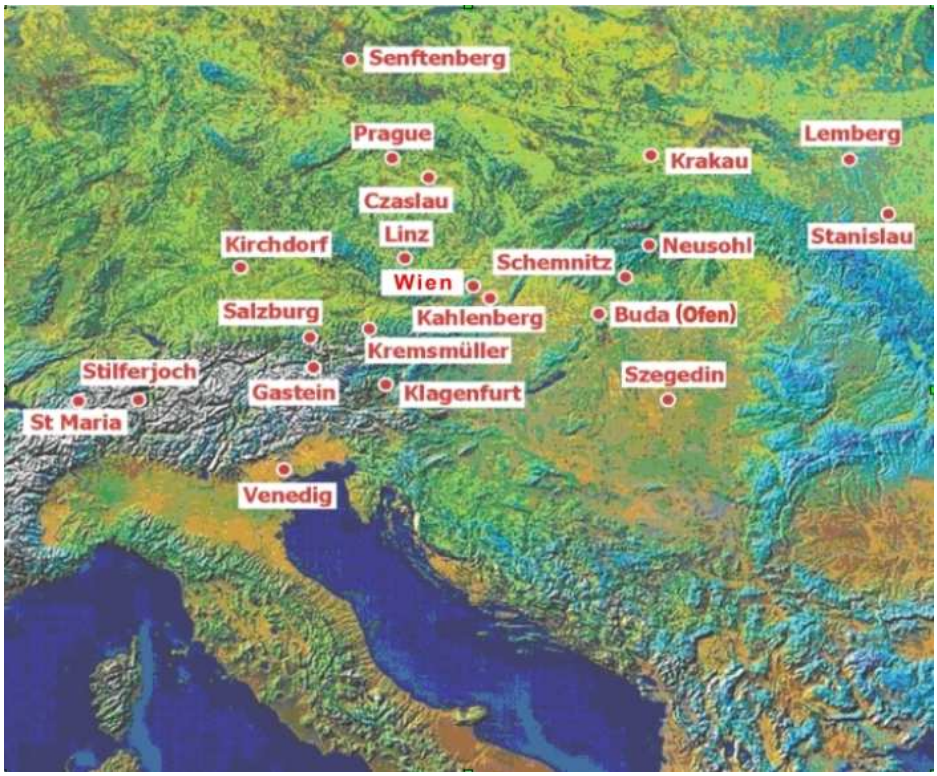

Figure 3. Ozone measurement sites in Central Europe between 1853–1856

Numerous meteorological stations in the Kingdom of Hungary also made ozone measurements using the Schönbein method (scale of 0–10) during the day and night: Buda (Ofen), Szeged (Szegedin), Selmecbánya (Schemnitz) and Besztercebánya (Neusohl). In this study, we will convert Schönbein numbers to ozone concentrations in ppbv using the approach outlined in Pavelin et al. (1999).

There are some interesting aspects of the measurement worth mentioning: (i) Vienna has quite a long ozone measurement period from 1853 to the end of 19th century; (ii) Ozone observation started in 1856 in Buda, however there was continuous published dataset in yearbooks only from 1871; (iii) in Krakau (Krakow), four Schönbein papers were simultaneously posted towards the four points of the compass to study the differences caused the wind speed outside the building (shielded vs. exposed); (iv) in Lemberg (Lviv) two daytime measurements were carried out; one in the morning and one in the afternoon. The concentrations measured were similar (Weidinger et al. 2009).

To calculate ozone level daytime and nighttime relative humidity data are needed. For humidity data missing in the yearbook, data from the nearest meteorological station were used. Of course it is a compromise but the selected stations were in the same climate region (Hungarian Plain). For Szeged we used humidity data from Temerin (near Belgrad) in one year, while in other years data was taken from Debrecen. In two other years measurements of humidity were found locally in Szeged. Meteorological measurements were usually carried out three times a day. There were two stations with more than three observations of humidity per day: in Seftenberg 5 times per day and in Kremsmünster 10 times per day. Daily (1 data/day) and monthly averages (for each daily measuring time) of relative humidity were published in the yearbooks.

3.2 Evaluation of ozone measurements in Szeged

Reconstruction of ozone data is illustrated on the dataset in Szeged (1853–1856). Schönbein's number showing annual variation with winter maxima and summer minima is shown in *Figure 4*.

Available dataset of relative humidity were i) daily averages (one value per day) and ii) monthly averages for three daily observations (0600, 1400 and 2000 hours in Central European Time). Monthly averages of relative humidity were determined by weighted averages of monthly values of three observations. Percentage differences between these values were used for reconstruction of daily variation of the relative humidity from each daily average. Using estimated relative humidity three times a day the daytime (between 0600–2000 hours) and nighttime (2000–0600 hours) relative humidity could be determined assuming linear changes between values.

Seasonal variability in daily average relative humidity is characterized by a winter maximum and a summer minimum (*Figure 4)*. In 1854 and 1856 extremely low daily average relative humidities were found in datasets from several stations suggesting these low values were not likely the result of measurement artifacts.

3.3 Spatial distribution of ozone over the Habsburg Empire in 1855

The good coverage of measuring sites of ozone allows to analyze the spatial distribution of ozone concentration. Density of measurements was quite high compared to today. The frequency of measurements allows us to determine daytime and nighttime ozone concentrations. The spatial distribution of ozone concentration in 1855 – produced from numerous measurements was characterized by maximum values around Vienna and low values (less than 10 ppbv) in Szeged and in Krakow (*Figure 5*).

3.4 Ozone deposition during the vegetation period over coniferous forests in Central Europe in 1855

A simple model using deposition velocities shown in *Table 1* was adapted to calculate ozone deposition over coniferous forests. The results for the vegetation growth season (from April to October) are presented in *Figure 6*.

Assuming the concentrations were very similar in daytime and nighttime, the differences between ozone deposition rates are caused by the different deposition velocities in daytime and nighttime (*Table 1*). Over coniferous forests during vegetation growth seasons ozone deposition was between 1,7 g/m^2 and 3 g/m^2. On the basis of land-use data from the 19th century (Timár et al. 2006, 2007) more precise calculations can be made.

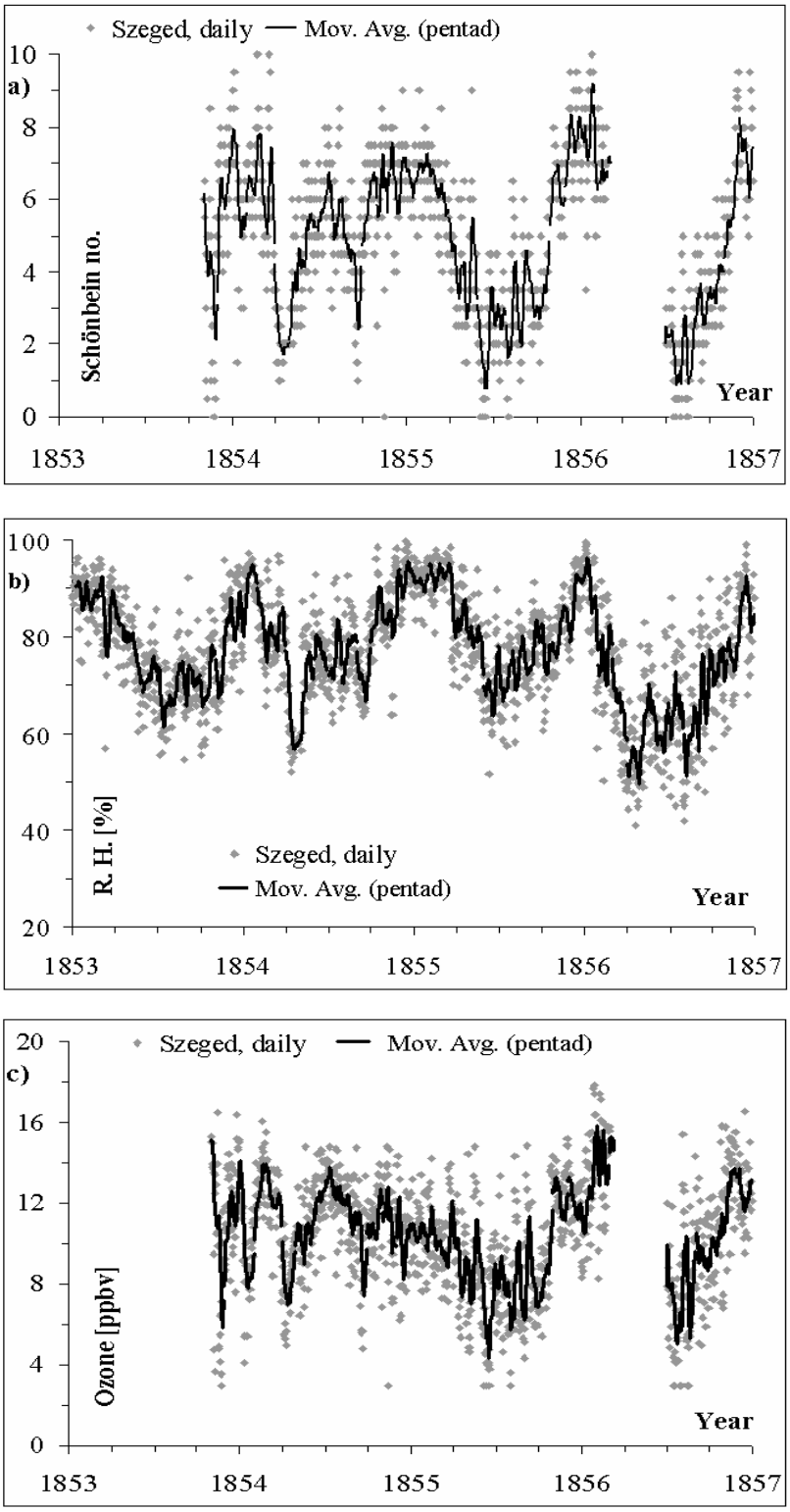

Figure 4. Schönbein number (a), relative humidity (b) and ozone concentration (c) in Szeged between 1853–1856. The moving averages were calculated over 10 days of data (pentad)

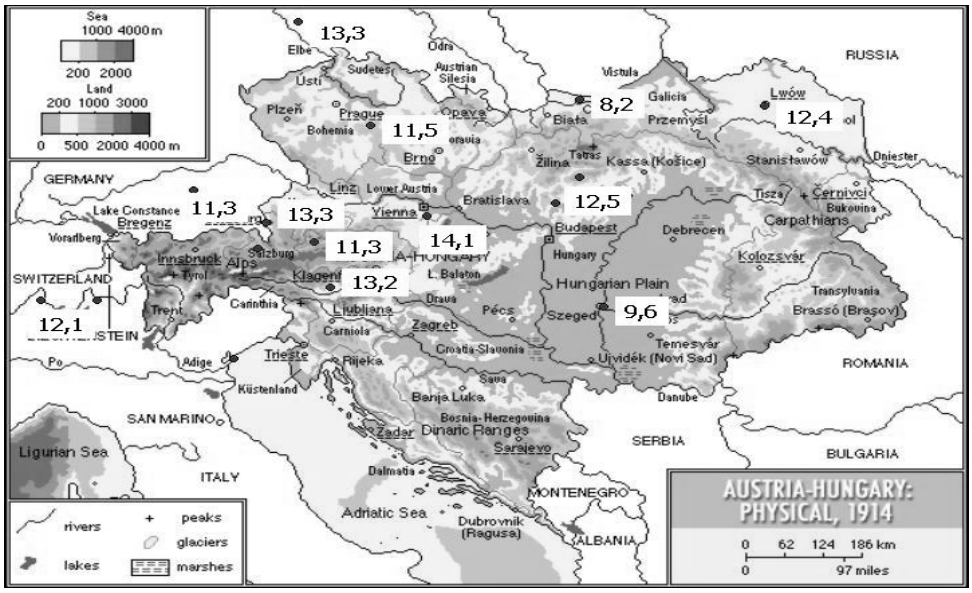

Figure 5. Annual average ozone concentration [ppbv] in the Habsburg Empire in 1855

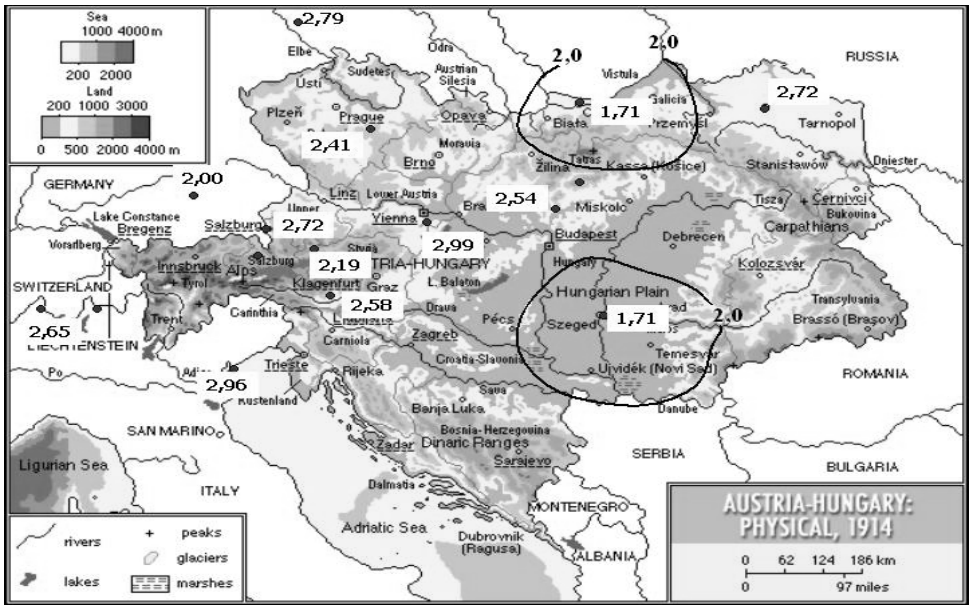

Figure 6. Ozone deposition [g/m^2] over coniferous forest during the vegetation growth season (April–October) on the Habsburg Empire in 1855, calculated on the basis of reconstructed ozone concentration and deposition velocity data measured during day- and nighttime in Nyírjes (Table 1) (The coniferous forest is an example. It is not autochthonous in the Hungarian Plain)

3.5 Ó-Gyalla Dataset (1893–1905)

The meteorological and geomagnetic observatory founded by Miklós Konkoly in Ó-Gyalla (now Hurbanovo, Slovakia) plays an important role in the history of Hungarian meteorology. Starting in 1900 it operated as the Observatory of the Meteorological and Geomagnetic Institute (Czelnai, 1995). Yearbooks of the Observatory contain ozone measurements (daytime and nighttime, on 0–10 Schönbein scale) besides meteorological and geomagnetic observations between 1897 and 1905. The relative humidity was measured three times a day similar to the former measurements at Szeged.

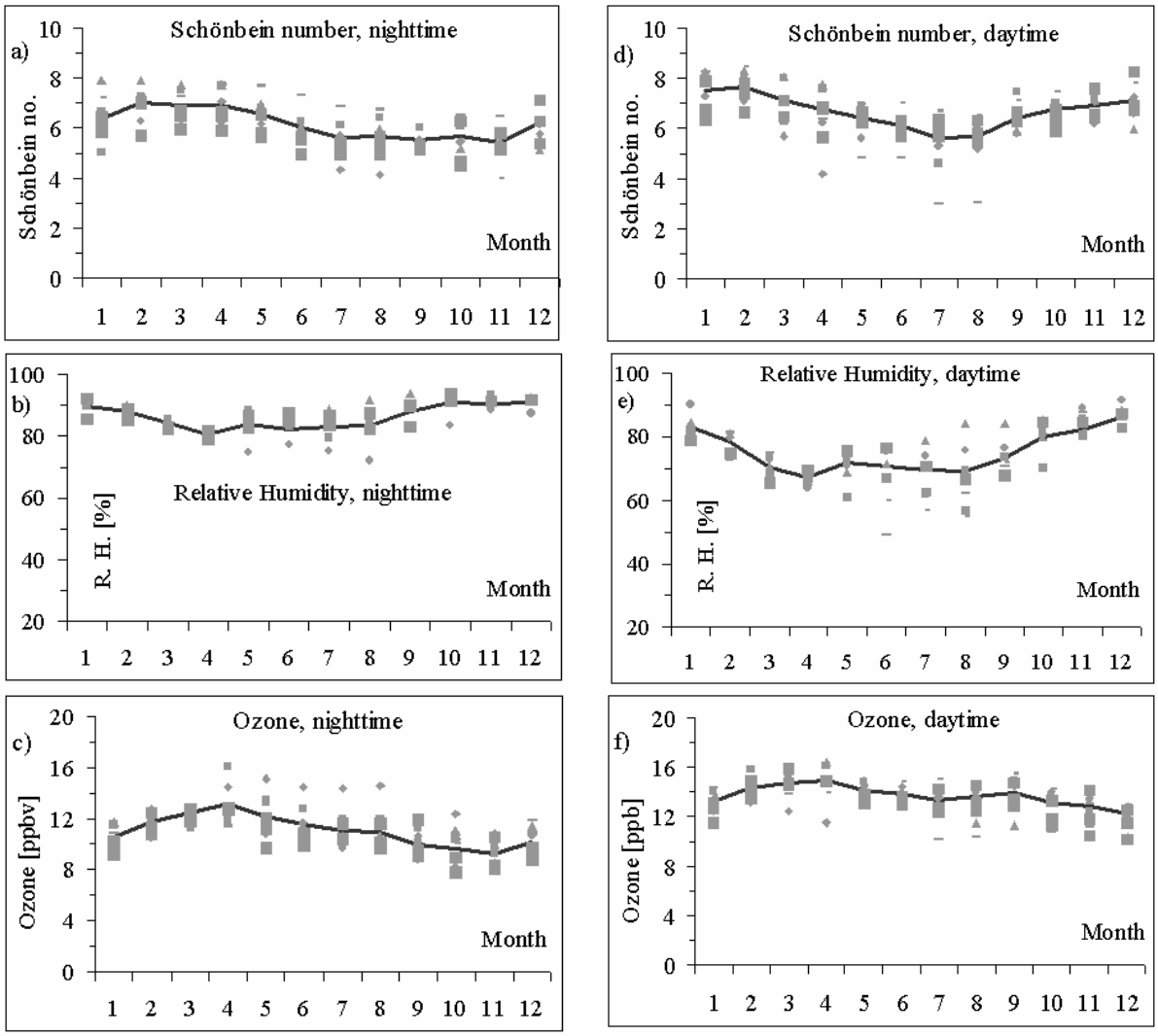

Figure 7. Monthly averages of nighttime and daytime Schönbein numbers (a, d), relative humidity (b, e) and ozone concentration (c, f) in Ó-Gyalla between 1893 and 1905

Figure 7 shows monthly averages for daytime and nighttime relative humidity, Schönbein number, and reconstructed ozone concentrations over a 13 year period. Average monthly values are indicated by continuous lines. Seasonal variation of ozone shows slight changes without summer maxima (not suitable anthropogenic effects compare with present). The daytime and nighttime average ozone concentrations – based on the Welch-test – are statistically different even at 99% significance level. Maximum values occurred in early spring (at daytime as well as nighttime), which are also found in the recent ozone dataset, explained by tropopause folding, which permits transfer to the surface (Sándor et al., 1994).

4 COMPARISON OF OZONE DATASETS AND FLUXES IN THE 19th AND 21st CENTURIES OVER URBAN AND RURAL AREAS

Daytime and nocturnal average ozone values are available for the stations located in the lower area of the Carpathian Basin for a period of over 150 years. In the evaluation ozone data measured at Szeged between 1854 and 1856, at Buda between 1871 and 1898 and at Ó-Gyalla between 1893 and 1905 were used.

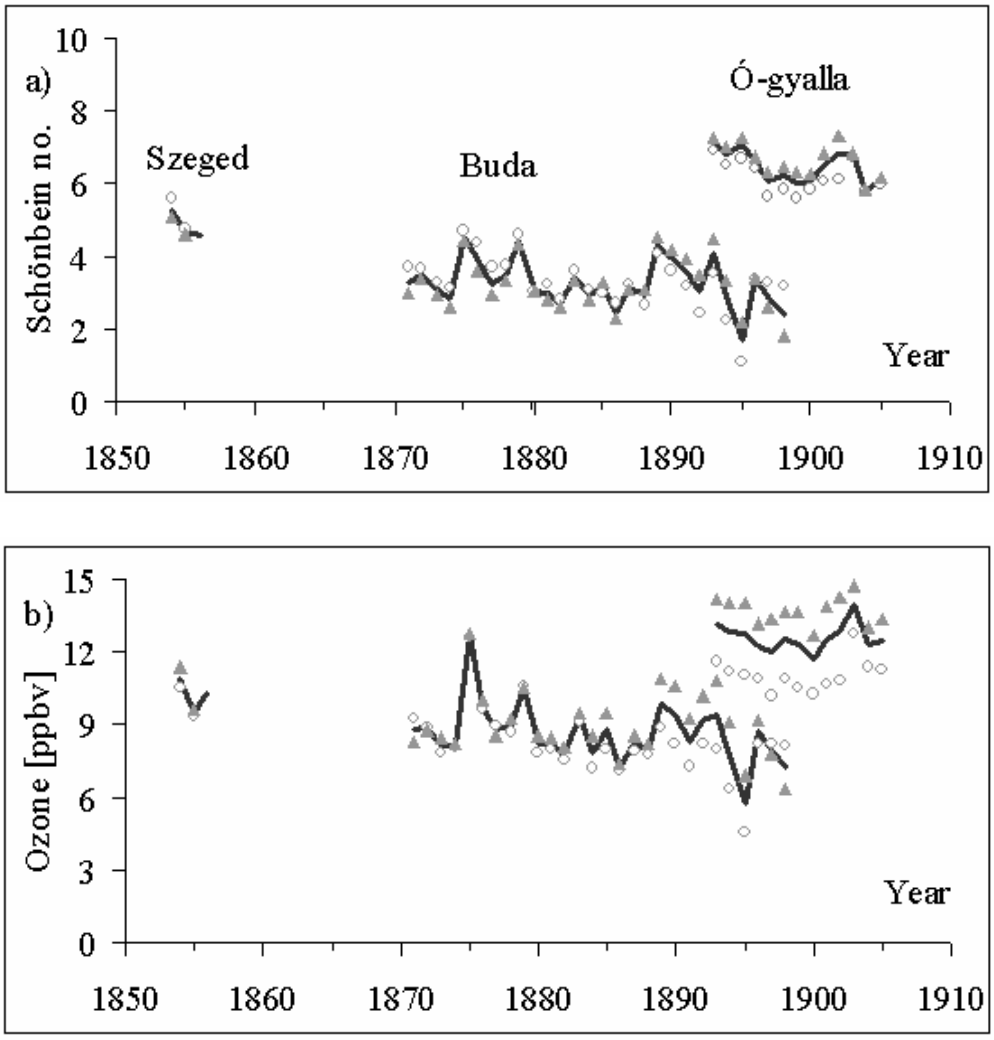

Figure 8. Annual averages of Schönbein number (a) and ozone concentration (b): daytime (grey triangles), nighttime (grey circles) and entire day (black line) measurements in Szeged, Ó-Gyalla and Buda between 1854–1904

The highest Schönbein value and ozone concentration are found in Ó-Gyalla and the lowest ones at Buda. The differences between ozone values at Buda and Szeged are very small. There are not significant trends in the ozone dataset in Buda. At the same time, measured ozone data in Ó-Gyalla (12–14 ppbv) and Buda (8–12 ppbv) indicate differences between urban and rural areas. This difference could be related to emissions of reducing gases such as SO_2 emitted in urban areas. Sulphate aerosol is formed in the atmosphere by the oxidation of sulphur dioxide. Ozone and the sulphate aerosol are interconnected to each other, since sulphate affects O_3 and the oxidant chemistry by providing a surface for the conversion of NO_x to nitric acid, limiting thus the formation of O_3 (Unger et al., 2006). On the other hand, reducing agents of the Schönbein paper – such as SO_2 and ammonia – can also lead to a lower ozone reading (Pavelin et al. 1999)

The daytime concentrations in spring and summer outpace in most cases the nighttime concentrations (*Figure 9*), which could be due to the effect of photochemical processes. In the majority of cases autumn and winter concentration at night is higher than in the daytime. Possible reasons are less daytime solar radiation, inactive period of vegetation

and the effect of the coal burning in could seasons. The tendencies are more pronounced in the summer and spring, while smaller changes are found in winter and autumn.

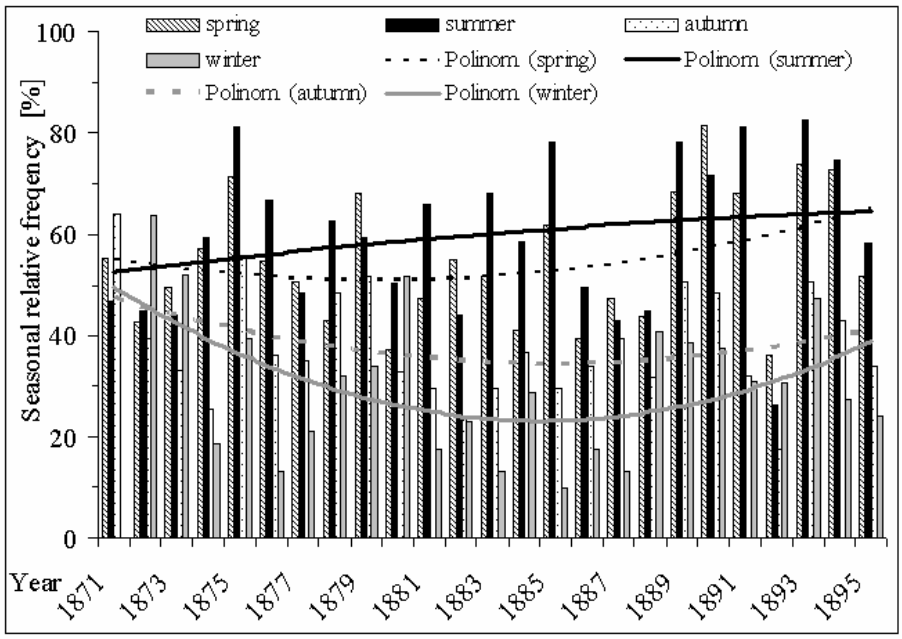

Figure 9. Relative frequency of days [%] when ozone concentrations are higher in the daytime (ΔC> 0) in different seasons observed in Buda between 1871 and 1895 (bars). Inter annual variation is demonstrated by fitted polynomial trend lines for each season (solid and dashed lines)

Taking into consideration of daily variation of ozone concentration from 1871 to 1895 in many days highest values were formed at nighttime, which are not common recently. For this reason these cases were treated separately. Long term variations of average differences were shown in *Figure 10* for winter and in *Figure 11* for summer. In winter smaller and in summer larger differences of concentrations can be found when the daytime concentrations are higher. It can be explained by photochemical reactions. Increasing trend can be found only in summer in this case.

The differences in ozone levels between urban and rural areas are increasing from beginning the observations until the present day. Differences between daytime and nighttime concentrations are also rising. Increasing emissions of nitrogen oxides and other trace gases playing important roles in ozone formation and the photochemical processes have been recognized (Mészáros 1997). Based on the measurements of the Hungarian Meteorological Service (for the period 1990–2008 as shown in *Figure 12*) increasing concentrations were found at both rural and urban areas until 1998. Since 1999 however, the concentrations are decreasing in both environments. Presently ozone levels are about three times higher than they were in the 19th century (compare *Figure 12* vs. *Figure 8*).

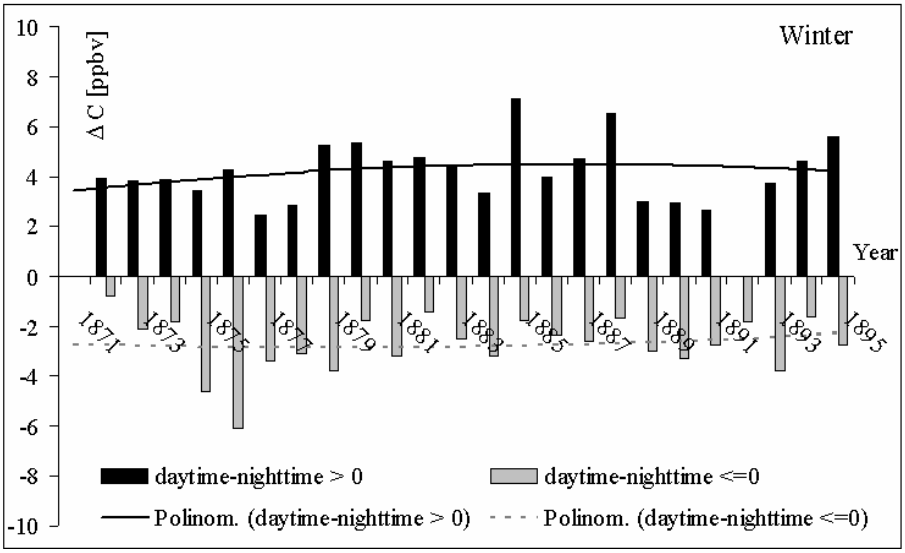

Figure 10. Average differences of daytime and nighttime measurements in Buda between 1871–1895 in winter for two cases i) for ozone concentration higher (dark) respectively ii) lower in daytime (gray). The variation between years is illustrated by trend lines

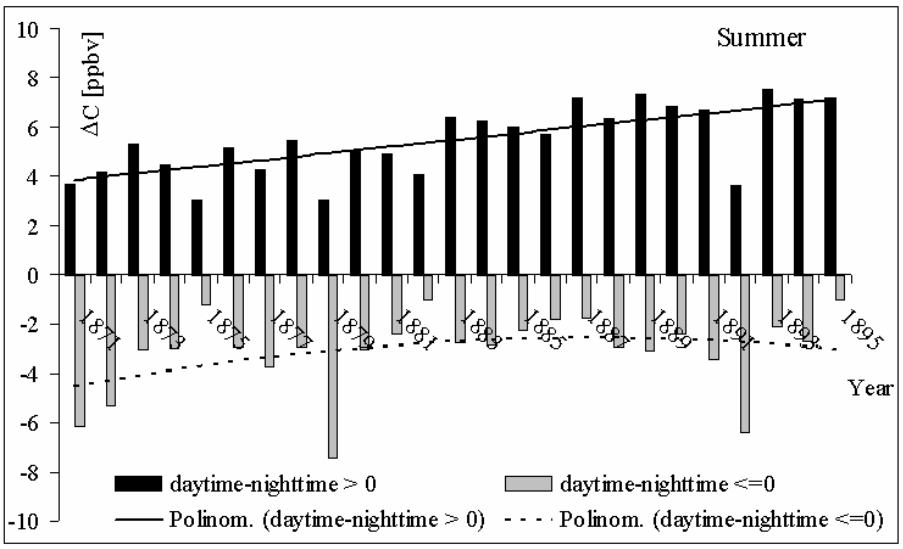

Figure 11. Average differences of daytime and nighttime measurements in Buda between 1871–1895 in summer for two cases i) for ozone concentration higher (dark) respectively ii) lower in daytime (gray). The variation between years is illustrated by trend lines

The change of ozone deposition from 19th to 21st century was illustrated over the coniferous forest for seven months (from April to October) of vegetation growth season *(Table 3).*

Table 3. Estimated ozone deposition [g/m^2] during the vegetation growth season (from April to October).

	Daytime	Nighttime	**Entire day**
Selmecbánya (1855)	2,2	0,3	**2,5**
Lemberg (1855)	2,4	0,3	**2,7**
Farkasfa (2008)	7,7	1,2	**8,9**
K-puszta (2008)	9,1	1,3	**10,4**
Nyírjes (2008)	9,5	2,0	**11,5**

Values almost three times higher were also found in later measurements cases than in earlier ones. In 1855, ozone deposition (*Figure 6.*) was 2.5 g/m^2 in Selmecbánya (Banska Štiavnica), while in Nyírjes 11.5 g/m^2 was calculated in 2008. Currently, differences of ozone deposition between two stations (Farkasfa and Nyírjes) are higher than the baseline deposition in the 19th century. In other words the spatial changes are larger than for the total deposition a century and a half ago. Due to anthropogenic activities, photochemical reactions play more important role at present. The differences between day and night values are increasing, and day to day variations in ozone concentrations are also increasing (see also Lagzi et al., 2004).

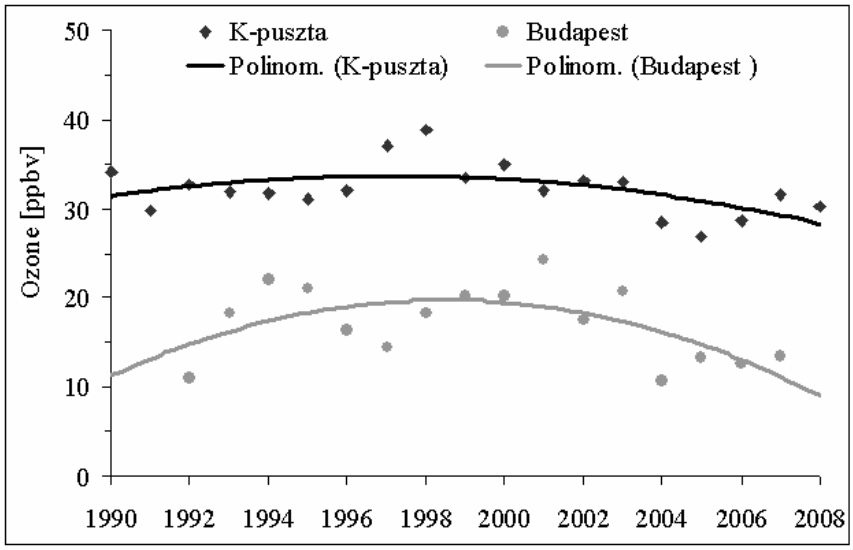

Figure 12. Annual averages of ozone levels between 1990 and 2008 in Budapest (urban site) and at K-puszta (rural site)

5 CONCLUSIONS

We present reconstructed ozone measurements and ozone deposition calculations based on these measurements (i) in the Habsburg Empire and in the Carpathian Basin in the middle of 19th century and the early 20th century; and (ii) in Hungary in the late 20th and the early 21st century. The Schönbein method is able to track the changes of concentration in the daytime and nighttime, and the seasonal changes as well. In summary:

- Reconstructed ozone concentrations in Central Europe are consistent with those deduced for measurements reported for other regions in the literature (i.e. generally in the range of 5 to 15 ppbv based on the widely accepted methodology of Pavelin et al. (1999).
- Although the data are subject to large uncertainties (~25% and possibly larger), they indicate that ozone concentrations were not likely to be substantially greater than this range and not as high as predicted by model calculations of pre-industrial background ozone.
- Small spatial variability of measurements over Central Europe between 1854 and 1905 for daytime, nighttime and daily values;
- Differences in ozone level between rural (Ó-Gyalla) and urban areas (Buda) were demonstrated;
- In Central Europe current ozone level and ozone deposition are about three times higher than in the 19th century.

Further investigations are planned on i) the effect of the wind speed, concentration of SO_2 and NH_3 for the ozone deposition according to the Schönbein paper, ii) uncertainty of the Schönbein method at high ozone concentrations and iii) the development of a multi-linear regressions model relating relative humidity and wind speed to Schönbein number based on the historical XIX century ozone measurements in Hungary.

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