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ORIGINAL PAPER

VARIABILITY AND TEMPORAL STRUCTURE OF CONCENTRATIONS OF CARBON MONOXIDE IN POZNAŃ (CENTRAL-WESTERN POLAND)

Eliza Kalbarczyk[1], Robert Kalbarczyk[2]

[1]Department of Spatial Econometrics
Adam Mickiewicz University in Poznan
[2]Institute of Landscape Architecture
Wroclaw University of Environmental and Life Sciences

Abstract

Carbon monoxide, in addition to CO_2 and CH_4, constitutes the largest source of carbon in the atmosphere and. by reacting with the hydroxyradical OH, it indirectly contributes to the increase in the concentration of methane and ozone in the atmosphere. The aim of the study was to determine the temporal variation of CO concentrations and its dependence on the weather course in an urban area situated in Wielkopolska, a region in central-western Poland. The research used data from the State Environmental Monitoring Station in the north-western part of Poznań (Hs = 84 m above sea level) in the immediate vicinity of a residential and recreational area. The data included hourly concentrations of CO and hourly values of atmospheric pressure, air temperature, total solar radiation, relative air humidity and wind velocity in 2005-2014. To measure the concentration of carbon monoxide, a Horiba APMA 360 analyzer and TELEDYNE API T400 were used. The measurement method was infrared spectroscopy. The time distribution of CO concentrations was considered by years, half years, months and hours. Relationships between the analyzed variables were confirmed using the Spearman's correlation coefficient at the level of $p \leq 0.01$. It was found that in the years of research the average concentration of CO in central-western Poland had a negative trend, as well as higher values in the heating season than in the warm half of year, and on week days than at weekends. Two concentration maxima, morning and evening ones, both in warm and cool half-year and on working days, were observed. On non-working days, there was one evening maximum. The variability of CO was mostly affected by air temperature (r_s = -0.45) and solar radiation (r_s = 0.32). The highest concentrations of CO were accompanied by the lowest values of solar radiation, air temperature and wind velocity, and highest values of the atmospheric pressure, as well as higher than average relative air humidity.

Keywords: gas pollution, meteorological factor, weekday, Wielkopolska region.

prof. UAM dr hab. Eliza Kalbarczyk, Department of Spatial Econometrics, Adam Mickiewicz University, Faculty of Geographical and Geological Sciences, Dzięgielowa 27, 61-680 Poznań, Poland, e-mail: ekalb@amu.edu.pl

INTRODUCTION

In the northern hemisphere, carbon monoxide is formed mainly during the incomplete combustion of fossil fuels and biomass (Granier et al. 1999, Novelli et al. 2003, John, Feyisayo 2013). It remains in the atmosphere for a relatively long time: on average for 3 months. The processes that remove CO from the atmosphere are primarily the reactions with hydroxyradical OH. Carbon monoxide can be transported over considerable distances, even across continents (Derwent et al. 1998, Newell, Evans 2000, Peng et al. 2007, Sahu et al. 2013). Although it does not directly influence the greenhouse effect, it contributes to the increase in the atmospheric concentration of greenhouse gases such as methane, carbon dioxide or tropospheric ozone (Derwent et al. 1998, Parrish et al. 1998, Peng et al. 2007, Rogalski et al. 2008, Worden et al. 2013, Lacressonnière et al. 2014). Tropospheric ozone arouses an interest of many researchers due to its toxic effects on plants and humans (Borowiak et al. 2011, Budka et al. 2014, Kalbarczyk et al. 2016). Also, the harmful impact of carbon monoxide on human health, although well known, is currently the reason for conducting numerous in-depth studies (Min et al. 2009, Du et al. 2010, Hampson 2011, Meyer et al. 2011, John, Feyisayo 2013).

The concentration of carbon monoxide, similar to methane, ozone and many other pollutants, is subject to distinct seasonal changes (Shahgedanova et al. 1999, Gorham et al. 2010, Kalbarczyk, Kalbarczyk 2010, Sahu et al. 2013, Rogalski et al. 2014, Kalbarczyk et al. 2015). The highest values are observed most frequently in winter or spring, while the lowest values occur in summer (Derwent et al. 1998, Parrish et al. 1998, Peng et al. 2007, Bigi, Harrison 2010, Laken, Shahbaz 2014). Generally higher concentrations are observed in the northern hemisphere than in the southern one (Granier et al. 1999, Laken, Shahbaz 2014). The average level of CO emissions in Poland, according to the Central Statistical Office of Poland (GUS) report (Enviroment Protection 2015), is ca. 280,000 tons per year, the largest source being non-industrial combustion (ca. 65%), 90% of which was of household origin. The second largest source was road transport (ca. 20% of the total carbon oxide emission). In terms of total emission, Poznań was the 20th on the list of Polish cities with a high environmental threat of emitting air pollutants from particularly noxious industrial plants (Enviroment Protection 2015). The average annual concentration of carbon monoxide in the Poznań agglomeration in 2014 was 324 μg m^{-3}, and it was nearly three-fold lower than concentrations in the most severely carbon monoxide contaminated city in Poland, i.e. Kraków (Enviroment Protection 2015). In Wielkopolska, according to the most recent data, no excessive levels of CO were found. In 2014, the 8-hour concentrations of CO in Poznań, depending on the location of measurement, amounted to 2560-2689 μg m^{-3}.

The concentration of CO shows distinct spatial variation, thus in places with high anthropogenic pressure the concentrations of carbon monoxide are

several times higher than the global average (Maffeis 1999, Meyer et al. 2011). Since the beginning of the 21st century, generally negative trends have been observed in CO concentrations over Europe, India, the United States and China, as well as in regional concentration growth areas, the largest ones being in China (Bigi, Harrison 2010, Worden et al. 2013, Laken, Shahbaz 2014, Yoon, Pozer 2014, Grechko et al. 2015).

The concentration of CO can be used as an indicator of carbon dioxide emissions of anthropogenic origin, formed during the combustion of fossil fuels (Chmura et al. 2010). The anthropogenic CO contribution is estimated to be from 83% in summer and 98% in winter (Mészáros et al. 2004).

The aim of the study was to determine the temporal variation of CO concentrations and its dependence on the weather course in an urban area.

MATERIAL AND METHODS

In the study we used data from a National Environment Monitoring station, located in the north-western part of Poznań. The station is situated on the grounds of a Botanical Garden (Hs = 84 m above sea level) in the immediate vicinity of a residential and recreational area. The measuring stand was located so that the air flow around the inlet and weather sensors was not limited by any obstacles. The sampling point was placed so as to ensure that the inlet does not suck again the air exhausted from the measuring station and the substances it contains before their sufficient mixing with the surrounding air. The height of the CO measurement point and the sensors to measure most of the meteorological features was 3 m above ground level, and only the sensors to measure wind velocity were at 10 m above ground level. Concentrations of carbon monoxide measured on the hour in the years 2005-2014 were analyzed. To measure the concentration of carbon monoxide, a Horiba APMA 360 analyzer and TELEDYNE API T400 were used. The method of measurement was infrared spectroscopy. Weather conditions were assessed on the basis of hourly meteorological data, which were: total solar radiation (Rad, W m^{-2}), air temperature (Ta, °C), relative air humidity (Rh, %), atmospheric pressure (Ph, hPa) and wind speed (WS, m s^{-1}), collected in the same multiple-year period as the data on gaseous air pollutants.

The temporal distribution of CO concentrations was considered by the years, seasons (cool half of year: October 1 – March 31, warm half of year: April 1 – September 30), months and hours. The average hourly data were used to determine the CO statistical indicators: the average ($\bar{x}$), maximum value (max), standard deviation (Sd), and the values of the lower (Q1) and upper (Q3) quartile. The incidence of the concentrations of analyzed pollutants in the preset ranges of values in the test period studied was also calculated. Daily data (calculated from at least 21 hourly measurements of the day) were used to determine the relationship between the concentration of

CO and the weather elements: Rad, Ta, Rh, Ph and Ws. Relationships between the analyzed variables were confirmed using the Spearman's correlation coefficient at the level of $p \leq 0.01$.

RESULTS AND DISCUSSION

The average annual level of CO concentrations in Poznań in the years 2005-2014 was 0.422 mg m^{-3} (Figure 1), and varied from an average of 0.484 mg m^{-3} in 2006 to 0.329 mg m^{-3} in 2012 (Figure 2). Values above the multi-year average were only in the first 5 years, while after the year 2010 all of the average annual values were lower than 0.422 mg m^{-3}. This confirms the general negative trend of CO concentrations in Europe in the 21st century

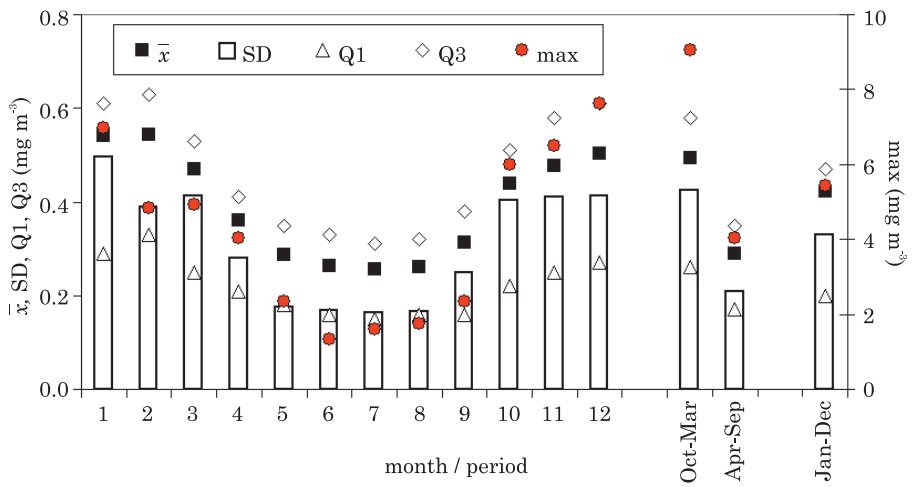

Fig. 1. Statistical characteristics: $\bar{x}$ – average, SD – standard deviation, Q1 – lower quartile, Q3 – upper quartile, max – maximum value describing the monthly, seasonal and annual variability of concentration of carbon monoxide. The years 2005-2014

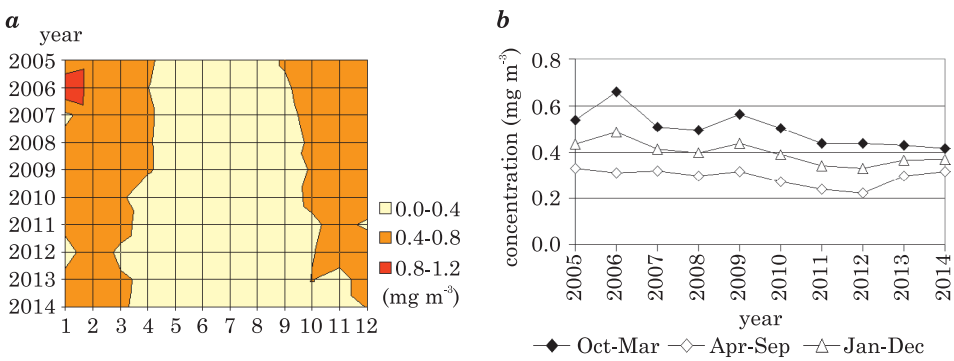

Fig. 2. Monthly (*a*), seasonal and annual (*b*) average concentrations of carbon monoxide in the multi-year period 2005-2014

(Worden et al. 2013, Laken, Shahbaz 2014, Yoon, Pozer 2014), also found in individual cases e.g. in Poznań (Merkisz et al. 2012), London (Bigi, Harrison 2010), or the Moscow region (Grechko et al. 2015). The reduction in the average annual CO concentration level happens mainly owing to a reduction of the concentration of this gas in the cool half-year. The average concentration in the heating season decreased in the research years by 0.24 mg m^{-3}, from 0.658 in 2006 to 0.415 mg m^{-3} in 2014 (Figure 2). In the warm half-year, with a generally lower level of CO concentrations, also the reduction in the average concentrations was smaller and amounted to ca. 0.11 from 0.329 mg m^{-3} in 2005 to 0.221 mg m^{-3} in 2012 (for both the periods it was thus about 60%).

As in other locations, not only European ones but also in South-East Asia (Kalbarczyk, Kalbarczyk 2010, Sahu et al. 2013), the seasonality of monthly concentrations of CO was pronounced (Figure 1). The highest average monthly concentrations of CO in Poznań were observed in February (0.544 mg m^{-3}), the highest maximum in December (7.630 mg m^{-3}), the lowest average in July (0.256 mg m^{-3}), the lowest of the maximum values in June (1.350 mg m^{-3}); in the cool half-year the average monthly and seasonal concentrations were approximately twice as high as in the warm half-year. The differences between the average monthly maximum values were much greater, the average maximum concentration of CO in December was more than 5 times greater than in June. Also the variability of the monthly concentrations expressed as standard deviation (Sd) was markedly greater in the cool half-year than in the warm half-year (Figure 1). The value of standard deviation of the monthly CO concentration in January (0.497 mg m^{-3}) was 2.5 times greater than the value of Sd in July (0.165 mg m^{-3}). In the cool half-year, the greatest standard deviation (0.626 mg m^{-3}) occurred in the year with the highest average concentration during this period, i.e. 2006, and was more than double the lowest one (0.296 mg m^{-3}), which occurred in 2013 (Figure 3). The average daily concentration of CO reached the highest values in the first days of January in the years observed (Figure 4a). The period of

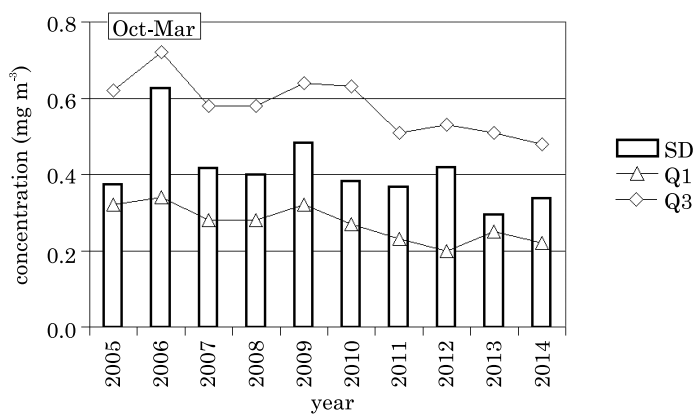

Fig. 3. Variation of the concentration of carbon monoxide in the successive years, described by the statistical indicators: SD, Q1 and Q3

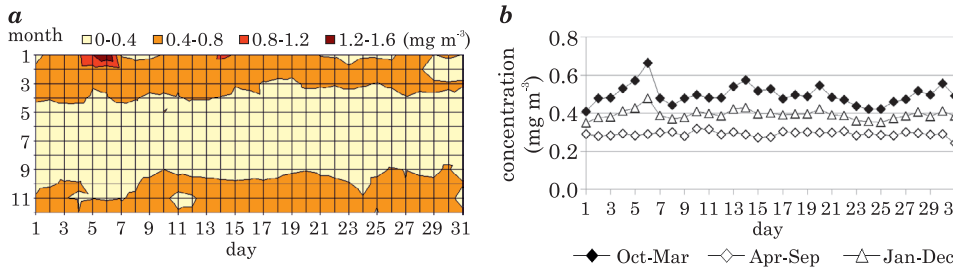

Fig. 4. Average daily concentrations of carbon monoxide by months (*a*), seasons and year (*b*). The years 2005-2014

elevated concentrations of CO (> 0.4 mg m^{-3}) was recorded on average from the second ten days of October to mid-April. Our analysis of daily average concentrations, according to half-years, again indicated a much greater variability of concentrations in the cold time of the year than in the warm half-year (Figure 4b). The average daily CO concentrations varied in individual months from 0.178 mg m^{-3} at the end of January to 1.729 mg m^{-3} in the first days of that month, while on average in the cool season (October - - March) they varied in the range from 0.407 to 0.663 mg m^{-3}. In the warm half-year, the range of variation was from 0.187 at the beginning of July to 0.467 mg m^{-3} in early April and on average throughout the warm season (Apr-Sep) from 0.272 to 0.318 mg m^{-3}. Markedly larger concentrations of CO in the cool season (winter or spring) and minima in summer were observed at many European measuring stations (Derwent et al. 1998, Meffeis 1999, Bigi, Harrison 2010, Laken, Shahbaz 2014), but also in northern China (Peng et al. 2007) or Canada (Granier et al. 1999). Different results were obtained in Moscow, where the highest concentrations were observed in spring and summer, together with the increase in traffic (Shahgedanova et al. 1999).

The CO concentrations also showed a marked daily variation, with two maxima (Figure 5). The highest average concentration of CO in a day in the cool half-year was usually in the early morning hours (6-8 h) and in the evening (19-21 h). Similar was the daily variability in the warm half-year, maximum values of on-the-hour concentrations occurred in the evening (20-21 h) and in the morning (ca. 6 h). Such variation is typical of locations with high traffic caused by commuting to and from work, and was recorded in London and Milan (Maffeis 1999, Bigi, Harrison 2010). In a study conducted in Los Angeles, the CO concentration reached its daily maximum in early afternoon, while the minimum was determined in late evening or early morning (Gorham et al. 2010). The smallest average hourly concentrations of CO in Poznań were most common in August at 12 to 16 hours (Figure 5a). The amplitude of change in concentrations in the heating season was not much greater than in the off-heating season, and was ca. 0.26 mg m^{-3}, from 0.367 to 0.632 mg m^{-3}; while in the warm half-year it was ca. 0.21 mg m^{-3}, from 0.205 to 0.414 mg m^{-3}.

In the course of the year and in the seasons considered, the most frequent,

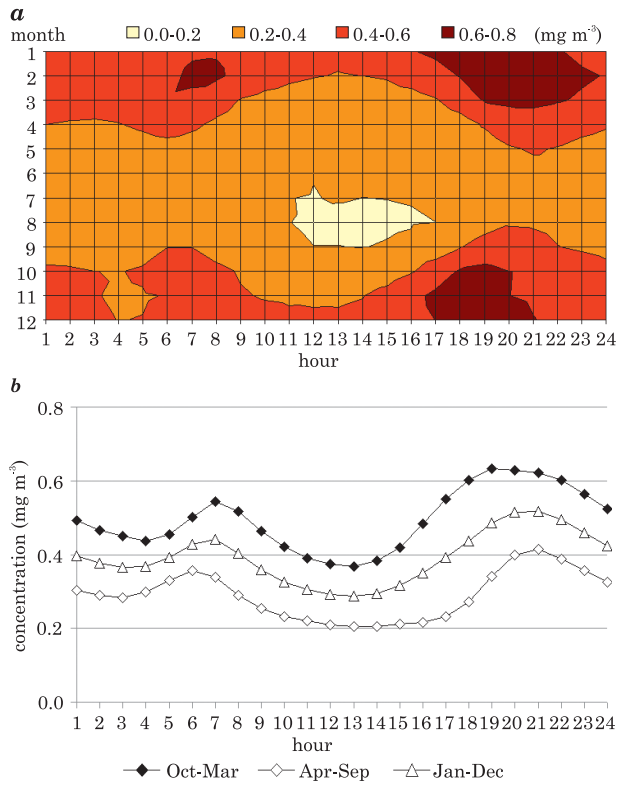

Fig. 5. Average hourly concentrations of carbon monoxide by months (*a*), seasons and year (*b*). The years 2005-2014

with frequency > 40%, were hourly concentrations in the range 0.2-0.4 mg m^{-3} (Figure 6). In the warm half-year, concentrations from the lowest concentration range, < 0.2 mg m^{-3} were measured with a similarly high incidence of 37%. In total, concentrations higher than 0.4 mg m^{-3} in the warm half-year

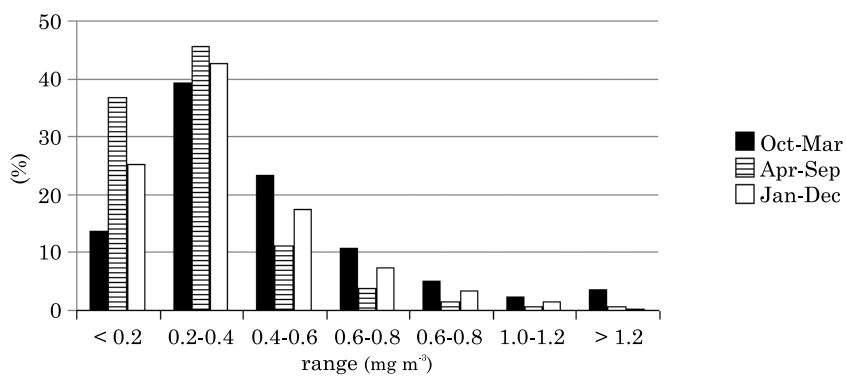

Fig. 6. Incidence of hourly measurements of carbon monoxide concentration according to seasons and year. The years 2005-2014

constituted more than 80% of cases. There were differences in relation to the cool half-year. With regard to incidence, the second most common was then the range 0.4-0.6 mg m^{-3}. The concentration range 0.1-0.6 mg m^{-3} in the cool half-year corresponded to > 60% of cases. In the range of highest values in the warm half-year, only fewer than 1% cases were observed, while in the cool half-year that percentage was ca. 3.5%.

Due to the dependence of CO concentrations on motor traffic (Janusz, Nadziakiewicz 2002, Drąg 2007, EEA 2015), their differentiation in the days of the week was also examined (Figure 7*a*). The common 5-day work week in Poznań resulted in two maxima of elevated concentrations in the days from Monday to Friday, with the highest values between 19 and 22 h, and slightly

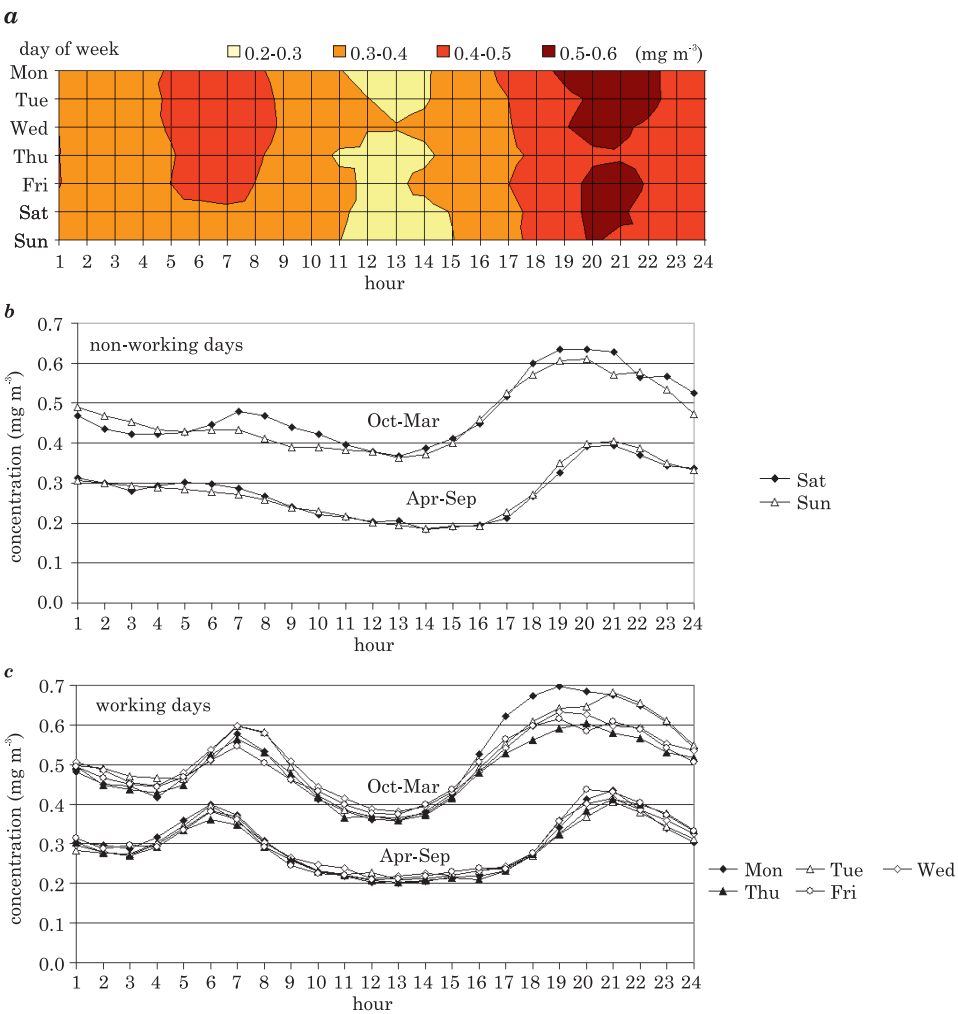

Fig. 7. Average hourly concentrations of carbon monoxide in the successive days of the week throughout the whole year (*a*) and in seasons (*b*, *c*). The years 2005-2014

lower ones at 6-7 h (Figure 7*c*), and a maximum on Saturday and Sunday evening at 20-21 h (Figure 7*b*). Rush hours in Poznań, according to data from 2013 (www. motofakty.pl), are 7:45-9:15 and 16-19. Thus Poznań, along with Warsaw, is one of the two large cities in Poland where peak traffic hours occur until evening. The most polluted day was the midweek Wednesday, when concentrations < 0.3 mg m^{-3} (Figure 7*a*) were not observed. In the remaining days of the week, concentrations < 0.3 mg m^{-3} were recorded for just 1 h (12-13 h) on Friday, up to 4 hours (11-15 h) on Sunday. A typical course for this type of concentration variation are highest values on Thursday and Friday, as a result of the accumulation of pollutants in working days (Maffeis 1999).

It is worth noting that the average level of CO concentrations on non -working days was just a little smaller than on weekdays. On non-working days it was in the range from 0.185 to 0.404 mg m^{-3} during the warm half--year, and from 0.364 to 0.635 mg m^{-3} in the cool half-year (Figure 7*bc*). On working days, it ranged from 0.201 to 0.438 mg m^{-3} during the warm half-year and from 0.359 to 0.698 mg m^{-3} in the cool half-year. In the case of CO measurements taken in inner cities, differences in concentration between the weekend and the other days are usually larger (Maffeis 1999, Bigi, Harrison 2010, Mapoma et al. 2014).

Maximum CO concentration values occurred in the cool half-year on Mondays, in the warm half-year on Fridays (Figure 7*c*), which most likely was caused by the frequent trips associated with weekend breaks outside of the city. Both on non-working days and working days, the level of concentration was markedly larger (about twice) during the cool half-year than in the warm half-year. This type of regularity can attest to the predominant influence of low emissions as the source of CO that impacts measurements at monitoring stations.

Variation in the concentration of carbon monoxide in fact depended on the variation in values of the meteorological elements. During the studied decade, the variation of air temperature was typical for Poland, the warmest was July, the coldest January (Figure 8). The sums of solar radiation reached highest values in May on average, the smallest increase was reported for December. December was the month with highest values of relative air humidity, which achieved its monthly minimum in June. In the winter months, the average wind speed was usually higher than the average, and the highest wind velocity throughout the year was observed in January, while the lowest one was in the period from July to October.

The highest value of te Spearman's correlation coefficient was calculated for carbon monoxide and average air temperature, then for CO and solar radiation (Table 1). Air temperature was the most important meteorological element affecting concentrations of CO in Mapoma's tests (2014). Increasing values of air temperature, solar radiation and wind velocity correlated with a decreasing concentration of CO. The negative relationship between the CO

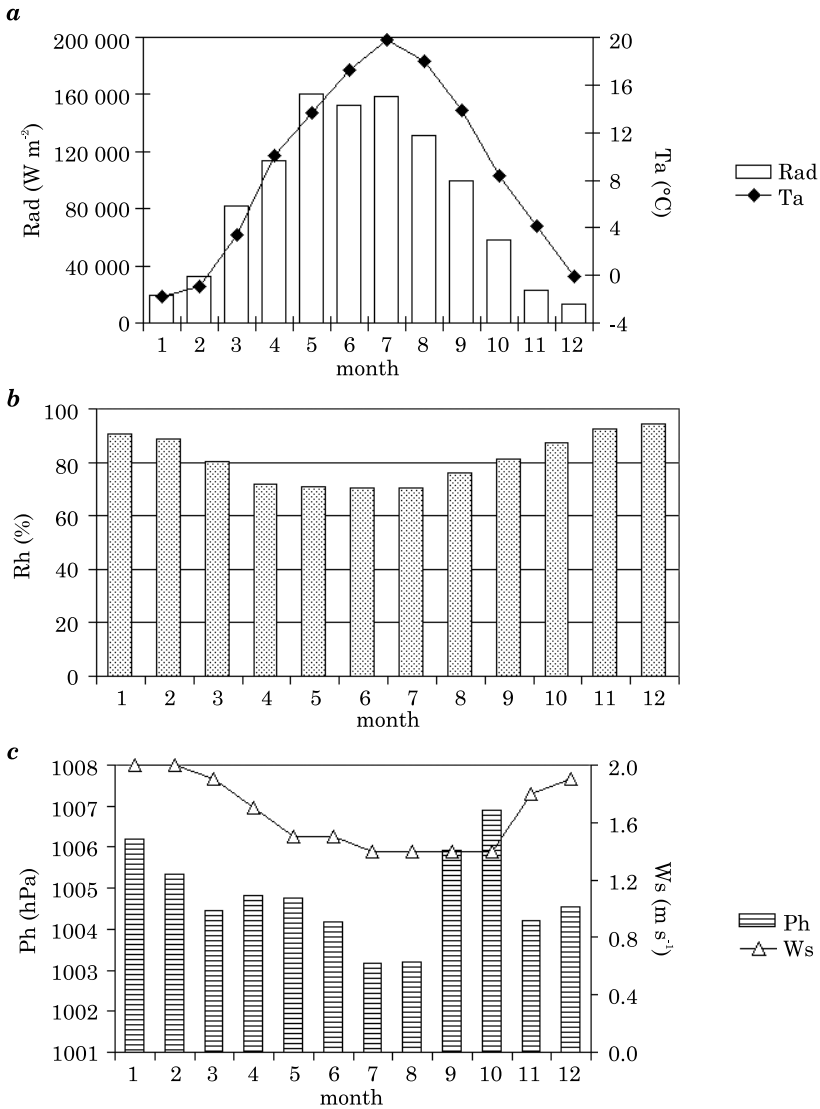

Fig. 8. Weather conditions described by the average hourly values of certain meteorological elements according to months in the years 2005-2014

concentration and wind velocity was reported by SHAHGEDANOVA et al. (1999). The direction and strength of wind was the major meteorological element used when modeling the spread of emissions from motor vehicles (JANUSZ, NADZIAKIEWICZ 2002). This relationship was positive only between relative air humidity and CO concentration. The conditions under which one can expect a higher than average concentration of CO are less-than-average values of the air temperature, solar radiation and wind velocity with simultaneously above-average values of relative air humidity. There was no relationship

Table 1

The dependence of the hourly concentration of carbon monoxide on annual meteorological elements and basic statistical characteristics describing their variability

Indicator	Meteorological element				
	Rad (W m^{-2})	Ta (°C)	Rh (%)	Ws (m s^{-1})	Ph (hPa)
r_s	-0.32	-0.45	0.26	-0.26	0.19
p	0.01	0.01	0.01	0.01	0.01
$\overline{x}$	123.7	8.9	80.9	1.6	1004.8
SD	201.4	9.3	20.1	1.1	8.7

r_s – Spearman's correlation coefficient, p – level of significance, x – average, SD – standard deviation

between the concentration of CO and the variation of atmospheric pressure, which was the most irregular weather component during the year compared with the other elements of the climate. The highest value of atmospheric pressure was recorded in October, the lowest was in July and also in August, when it was on a very similar, low level. However, correlations between the CO concentration and atmospheric pressure have been demonstrated by Ćwiek and Majewski (2015), and a higher level of the CO concentration on days with low wind velocity and high pressure system (anticyclone) was reported by Maffeis (1999).

Our analysis of the average hourly concentrations in the designated intervals against the background of the course of monitred meteorological elements indicated a similar CO concentration and atmospheric pressure value system, opposite to average hourly amounts of radiation, the average air temperature and wind velocity. The highest concentrations of CO (> 1.6 mg m^{-3}) were accompanied by the lowest values of solar radiation, air temperature and wind velocity, maximum atmospheric pressure values, as well as higher than average air humidity (Figure 9). This last element, i.e. relative humidity, changed clearly only at lower CO concentrations (0.1-0.6 mg m^{-3}), whereas at medium and high levels of O concentrations, the values of relative humidity hardly differed between each other.

CONCLUSIONS

Concentrations of CO in built-up areas in the years 2005-2014 had a negative trend and distinct seasonality. The highest concentrations were reported in the cool half-year, i.e. in the heating season. The most polluted month in a year was February, the lowest average concentration was reported for July, the average concentration in the cool half-year and the winter

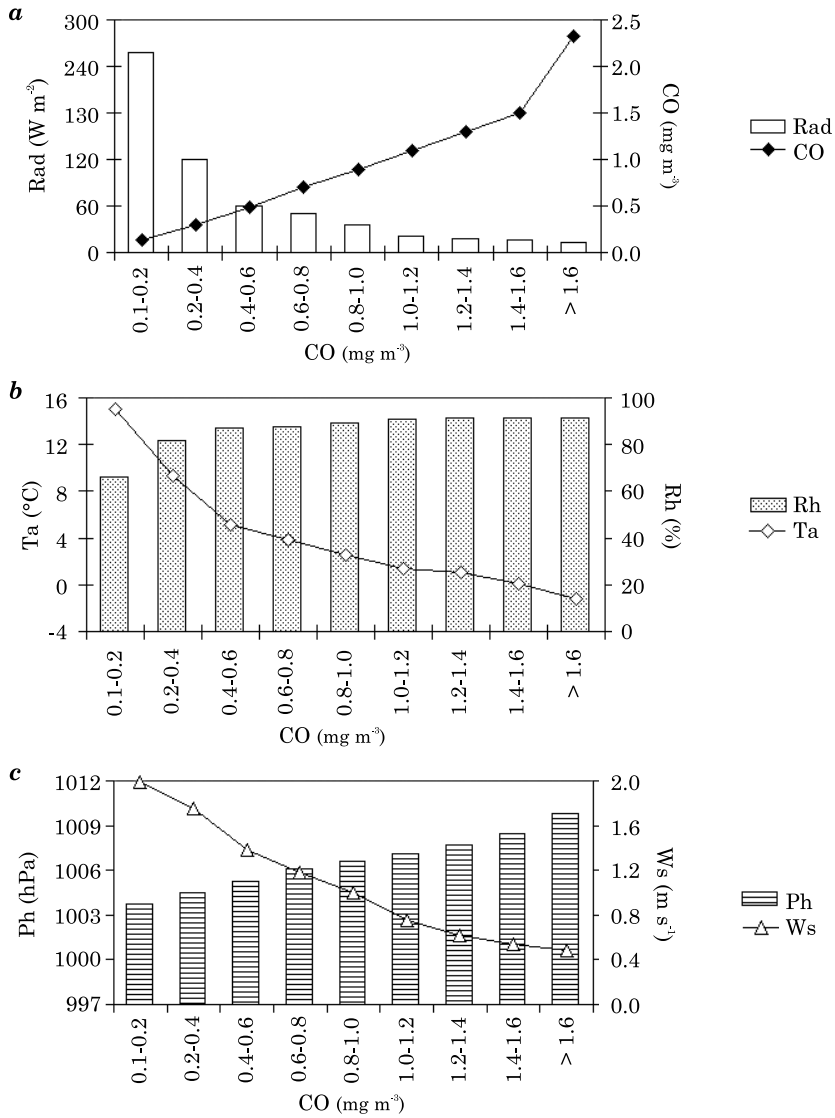

Fig. 9. Average hourly values of carbon monoxide concentration calculated at designated intervals along with meteorological conditions. The years 2005-2014

months was approximately twice as high as in warm periods of the year. The most frequently observed CO concentration ranged between 0-40 mg m^{-3}. Throughout the day, there were usually two maxima, morning and evening ones, both in warm and cool half-year, and on working days; on non-working days, there was one evening maximum.

The CO concentration was lower at weekends than on weekdays, although the differences were relatively small. Variation in the concentration of

CO depended on the variation of weather conditions, primarily air temperature, but also solar radiation and relative air humidity. Our analysis of the variation in CO concentrations pointed to the dominant influence of low emission on the course of CO concentrations.

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