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Observation of Unusual High Particulate Mass and Number Concentration during Traffic Ban Hours of the 2009 Car Free Sunday in the Brussels Urban Area

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Abstract

Every year, since 2002, the Brussels authorities organize a car free day on the third Sunday of September. This very interesting experience has revealed some valuable information concerning traffic-related gaseous pollutants and particulates. On the car free Sunday of 2006 very high PM10 and PM2.5 mass concentrations were measured, along with very low concentrations for nitrogen oxides, carbon monoxide and dioxide. The car free Sunday of 2009 also showed very interesting results. During the traffic ban hours, particulate mass concentration and particulate number concentration peaked to one of the highest values for the whole year 2009. Black Carbon however was the only measured particulate component whose concentration continued to decrease during the traffic ban period.

Keywords: *car free Sunday, PM10, PM2.5, black carbon, particle mass concentration, particle number concentration*

Rezumat. Observaţii asupra masei şi concentraţiei neobişnuit de mari ale particulelor în suspensie înregistrate în duminica fără trafic a anului 2009, în aglomeraţia urbană Bruxelles

Începând cu 2002, autorităţile bruxeleze au instituit în fiecare an o zi în care traficul este interzis, în cea de-a treia duminică din luna septembrie. Această experienţă foarte interesantă a pus în evidenţă informaţii valoroase privind sursele gazoase de poluare, consecinţă a traficului intens şi particulele în suspensie. În duminica fără trafic a anului 2006, au fost înregistrate concentraţii extreme ale PM10 şi PM2,5, împreună cu concentraţii scăzute ale oxizilor de azot, monoxidului şi bioxidului de carbon. Duminica fără trafic a anului 2009 a evidenţiat, de asemenea, rezultate interesante. Pe parcursul orelor cu trafic interzis, masa şi concentraţia numărului de particule în suspensie au atins valori dintre cele mai ridicate din 2009. Carbonul elementar (funinginea) a rămas singurul component măsurat a cărui concentraţie a continuat să descrească pe parcursul perioadei de trafic interzis.

Cuvinte-cheie: *duminica fără trafic, PM10, PM2,5, carbon elementar (funingine), masa particulelor, concentraţia particulelor*

GENERAL SITUATION AND METHODOLOGY

The air pollution telemetric network in the Brussels Capital Region consists of eleven fixed stations, located in different types of urban environment (traffic, residential, industrial and urban background), measuring a selection of several pollutants: nitrogen monoxide and dioxide, ozone, sulfur dioxide, carbon monoxide, carbon dioxide, benzene, mercury vapor and the PM10 and PM2.5 mass fraction of the suspended particulates (particulates with an equivalent diameter up to 10 or 2.5 μm). The presence of nitrogen oxides is monitored in all 11 measuring sites, PM10 in 6 locations and the PM2.5 mass concentration is

simultaneously measured in five of the six PM10 locations. PM10 and PM2.5 mass concentrations are measured by means of TEOM 1400Ab continuous instruments (*Tapered Element Oscillating Microbalance*), all equipped with FDMS 8500 modules (*Filter Dynamics Measurement System*). Nitrogen oxides are measured by ThermoFisher NO_X-analyzers, model 42C or 42i.

The EU air quality objectives from Directive 2008/50/EC are respected for most pollutants in most of the Brussels measuring sites. However, full compliance is still problematic for NO_2 at traffic oriented sites and for PM10 at sites along the industrial and economic axis of the Region. The NO_2 limit value, 40 µg/m^3 as the annual average, is to be respected from 2010 and since 2005 the PM10 daily limit value of 50 µg/m^3 may not be exceeded more than 35 times per year. Observations of the average NO_2 concentrations for Saturdays and Sundays and of the number of PM10 exceedings during the weekend, make clear that emission reductions, even as drastic as lowering the every day emissions to the actual average weekend emissions, will not enable to meet the air quality objectives in all Brussels measuring sites (Vanderstraeten, 2010b).

Over the past few years (2004-2010) the annual average NO_2 concentration level reached about 50 to 55 µg/m^3 in a street canyon, about 42 to 46 µg/m^3 in sites exposed to the traffic but situated in a better ventilated area, about 37 to 42 µg/m^3 in urban sites and 27 to 30 µg/m^3 in residential or urban background sites. On Saturdays the mean concentration is around 48 to 52 µg/m^3 in the street canyon, about 38 to 42 µg/m^3 in the exposed sites and about 24 to 28 µg/m^3 in the urban background sites. On Sundays the mean concentration is around 40 to 42 µg/m^3 in the street canyon, about 30 to 35 µg/m^3 in the exposed sites and about 20 to 24 µg/m^3 in the urban background sites (IBGE technical report, 2009a).

Over the past decade (1999-2010) a slow downward tendency of PM10 levels was observed. The PM10 annual average at the different sites in the Brussels area ranges from about 27 to 30 µg/m^3 for the urban background and suburban site to 30-35 µg/m^3 at the traffic site along the economic axis, to hardly less than 40 µg/m^3 at the industrial site of the naval Port. Although the EU limit value for the annual PM10 average concentration (40 µg/m^3) was respected in all Brussels sites since 2005, the limit value for the daily average concentration was systematically violated in two sites situated along the industrial and commercial axis, and occasionally, this was also the case at some of the other sites. For the years 2007 and 2009, due to several periods of poor dispersion during the winter months and the frequently observed formation of secondary aerosol during the spring, at the start of the agricultural season, this limit value was respected only in one, respectively in two of the six PM10 sites.

Since June 2008, at the Woluwe traffic site situated along a highway leading traffic into Brussels, the particulate number concentration is also measured, for 31 different particulate classes with equivalent diameters ranging from 0.25 µm to 32 µm, by means of a Grimm Laser light scattering spectrometer model 365. Since July 2009, at the same site, the mass concentration of 'Black Carbon' is measured by means of an aethalometer, Magee Scientific model AE22-ER. Over the past years air pollution by particles, the role of traffic, source appointment and the chemical composition were subject to numerous studies in Europe and the USA (Almeida et al., 2006; Hansen et al., 2010; Harrison et al., 2001; Holmes et al., 2007; Ruuskanen et al., 2001).

TRAFFIC RELATED GASEOUS POLLUTANT LEVELS DURING CAR FREE SUNDAYS

On all previous car free Sundays (2002-2008) a clear concentration decrease was observed for traffic related gaseous pollutants, but this was generally not the case for the PM10 and PM2.5 mass concentration. Unlike for the gaseous pollutants, relatively high PM mass concentrations were measured during the 2003 car free Sunday and in 2006 the daily PM10 limit value was even exceeded (Vanderstraeten, 2010a).

On Sunday, 20 September 2009, for the eighth time since 2002, a car free Sunday was organized by the Brussels Authorities. From 7:00 till 17:00 h UT (UT: Universal time or GMT), corresponding to 9:00 till 19:00 h local time in the summer, nearly all motorized traffic was banned on the roads over the entire Brussels Capital Region. The use of Public transport was free and besides the busses from the Brussels Public transport company, exceptions were given to a limited number of taxis, to emergency services and, on request, to a few thousands individuals. During the traffic ban hours the speed limit was set at 30 km/h.

The day was characterized by mild meteorological conditions: a temperature inversion close to the surface from midnight until 7:00 h UT, rather high relative humidity (85 to 90%) in the morning which decreased towards the afternoon (60 to 70%), weak wind (1-2 m/sec) until 9:00 h UT

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and moderate wind (2-3 m/sec) in the afternoon, with the temperature rising from 15°C in the early morning to about 21°C in the afternoon. Between midnight and 8:00 h UT the wind was mainly coming from the South, but then suddenly it changed direction and began blowing from north-northwest until late in the evening, importing polluted air masses with secondary aerosols already formed over the western part of Belgium. In Brussels the clear morning sky was replaced by a thin cloud cover and a reduced visibility for the rest of the day. The boundary layer height computed from ECMWF (*European Centre for Medium Range Forecast*) fields, was about 60 m until 6:00 h UT, then it increased progressively during the morning to reach about 950 m in the afternoon. Therefore, the meteorological conditions can be considered as relatively unfavorable to pollutant dispersion during the night and the early morning, but rather favorable during the late morning and the afternoon.

The results of the traffic ban can be read easily from the graphs representing the concentration evolution on the car free Sunday, an average Sunday and an average working day during the period May–September 2009 (Fig. 1). The concentration decrease is best seen in a road tunnel where the concentration levels are much higher than in the ambient air and where the influence of the meteorological conditions on the concentration is less important. Figure 1 represents the concentration of NO in a road tunnel leading to the centre of the city. Before and after the traffic ban hours the concentration on the car free Sunday (in the front graph) follows that of the average Sunday. A sharp and sudden decrease of the concentrations can be observed at 5:30 h UT, even before the official start of the traffic ban period. By the end of that period (17:00 h UT), when the traffic returns, a sudden increase of the concentrations can be seen. Similar trends are also obtained for NO_2 and CO, for any of the car free Sundays and for the average of all eight car free Sundays so far organized.

Similar, but less striking observations could be made in the past, for the traffic-related gaseous pollutants at traffic oriented ambient stations. Since the ambient concentrations on one particular day are strongly dependent on meteorological conditions, it is more appropriate to represent the average situation over all eight car free Sundays and to compare it to the average situation of all Sundays and all working days during the period May–September of the years 2002-2009, as illustrated in Figure 2. For this traffic site, similar trends are obtained for the other traffic-related gaseous pollutants NO, CO and CO_2.

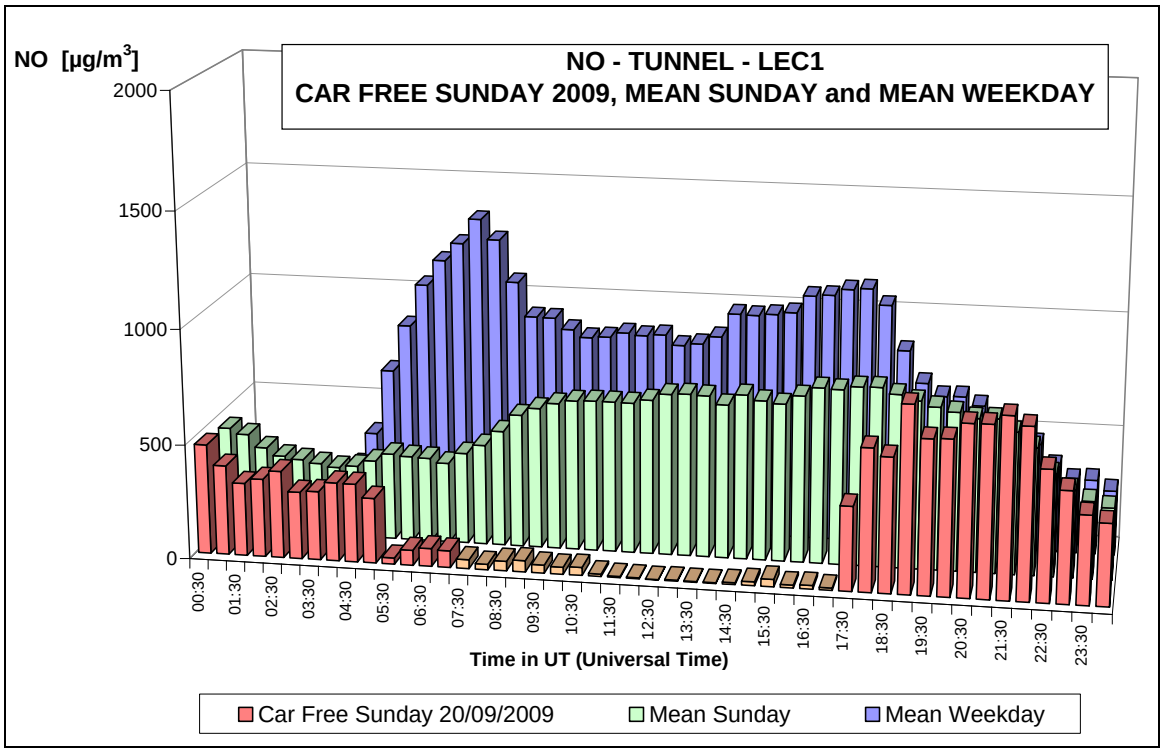

Fig. 1. Road Tunnel NO concentration – Car Free Sunday 20/09/2009, average Sunday and average working day during 'May-September 2009'

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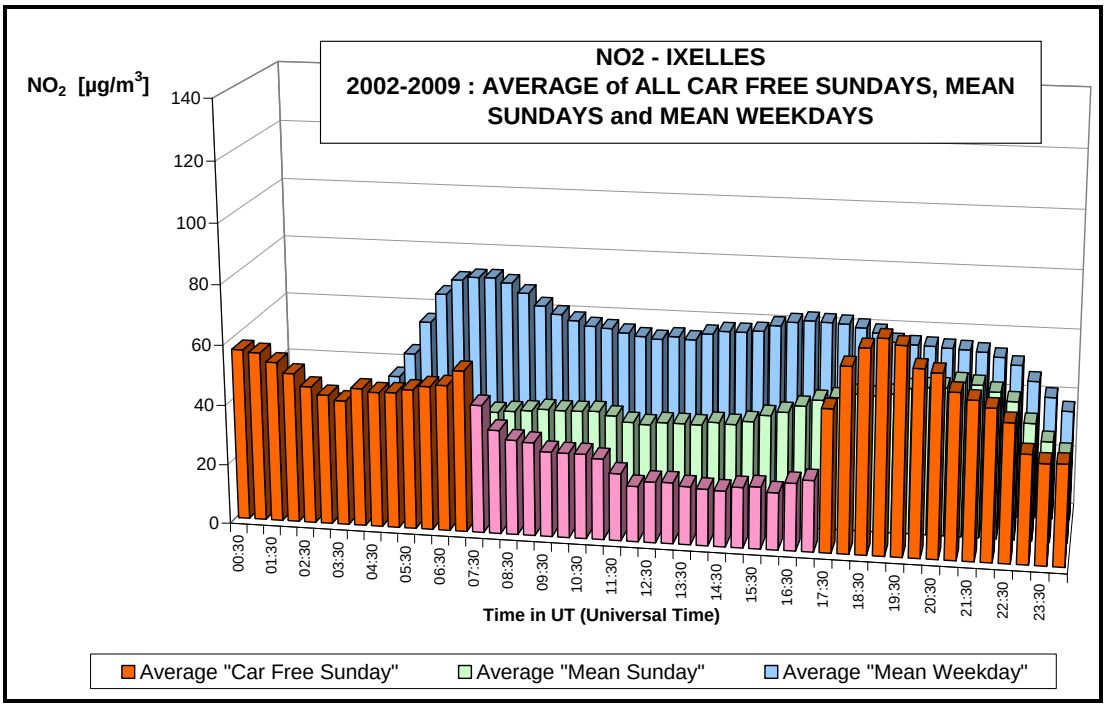

Fig. 2. NO_2 at a traffic oriented site - Average of all eight car free Sundays (2002-2009), average Sunday and average working day during May – September 2002-2009

In spite of the dependency on the prevailing meteorological conditions, the concentrations of the traffic-related gaseous pollutants were always found to be lower during the traffic ban period compared to the same period on an average Sunday or average working day, with an exception for the short peak at noon during the 2009 car free Sunday. A sudden concentration change always appeared at the beginning and at the end of the car free period with a clear sink of the concentration in between. Furthermore, a decrease of the NO_2 concentration was observed in all eleven Brussels measuring sites, in traffic oriented as well as in urban background and suburban sites (IBGE technical report, 2009b). This observation makes clear that the NO_2 problem with respect to the EU limit value could be solved if local traffic NO_X emissions were to be reduced drastically.

PM10, PM2.5 and Black Carbon during the Car Free Sunday

The PM10 and PM2.5 concentrations on the car free Sunday of 20 September 2009 were higher than on an average Sunday or average working day during the period May–September 2009. Most surprising however was the sharp concentration increase during the traffic ban hours, peaking at around 10:00h UT, followed by slowly decreasing but still elevated concentrations until the end of the interdiction time. This can be read from the Figures 3 and 4 representing, for the Molenbeek site, the concentration evolution respectively for PM10 and PM2.5 on an average Sunday, an average working day and the 2009 car free Sunday. For the readability of the graph the high concentration levels of the car free Sunday are moved to the back. Although the individual half hourly values of the car free Sunday are compared to computed average values for the mean Sunday and working day, these values are exceptional high.

Similar results were obtained in all Brussels sites measuring PM10 or PM2.5. The car free average daily PM10 and PM2.5 values respectively ranged from 47 to 64 µg/m^3 and from 38 to 52 µg/m^3 with daily PM10 averages exceeding the 50 µg/m^3 limit value in five of the six PM10 sites. On a total of eight car free Sundays, the daily PM10 limit value was exceeded twice, in 2006 and 2009, and it failed to do so in 2003. During the interdiction hours, the PM10 and PM2.5 levels were about two times higher than during the rest of the day: the traffic ban average concentration at the different sites ranged from 66 to 93 µg/m^3 for PM10 and from 58 to 76 µg/m^3 for PM2.5. As it was the case during the 2006 car free Sunday, the PM2.5 mass concentration represented about 80 to 90% of the total PM10 mass concentration. For all stations, the maximum half hourly values measured during the traffic ban hours, were ranging between the 99.3 and 99.8^{the} percentile of the whole year 2009 (IBGE technical report, 2009b).

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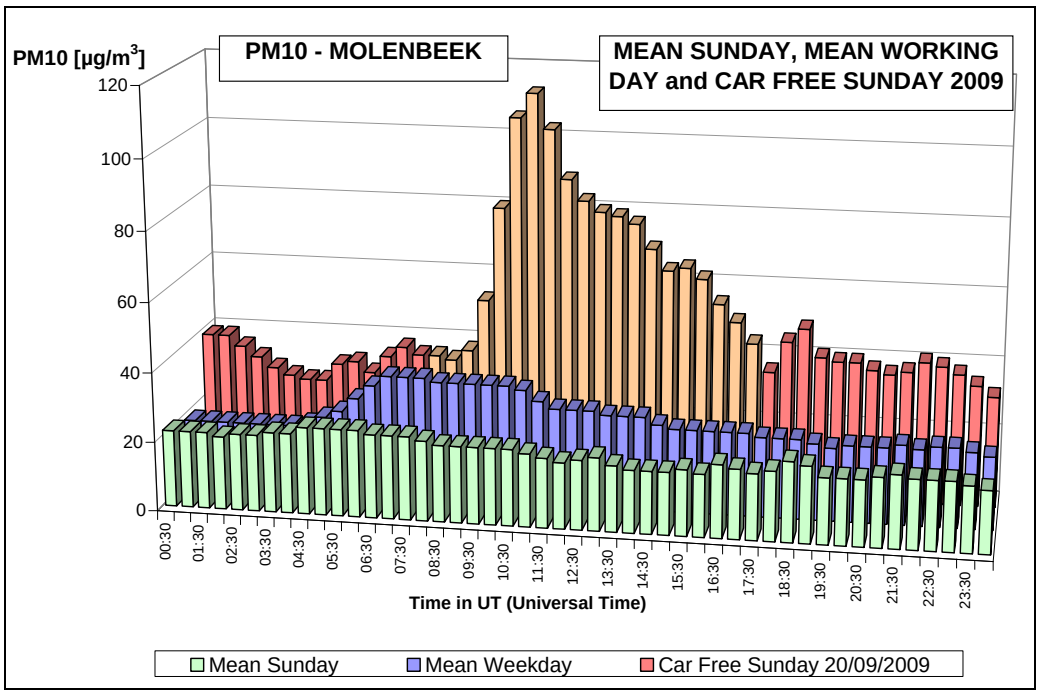

Fig. 3. PM10 mass concentration at Molenbeek - Average Sunday and average working day during 'May-September 2009' and Car Free Sunday 20/09/2009

At the beginning and at the end of the car free period one cannot observe a sharp or sudden concentration change, no concentration drop at the start nor a concentration increase at the end of the traffic free period, as is the case for traffic related gaseous pollutants such as NO, NO_2, CO and CO_2. These very high concentrations were obtained despite the absence of motorized traffic during that part of the day, with only minor contributions of domestic heating (15 to 21°C ambient temperature) and restricted industrial and commercial activities (Sunday).

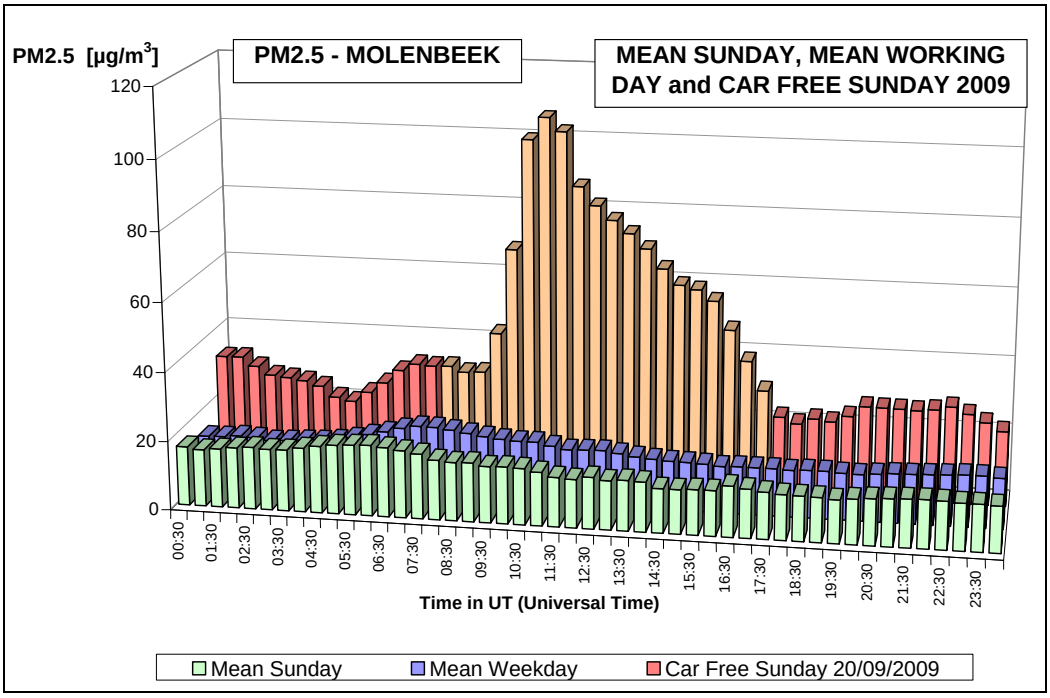

Fig. 4. PM2.5 mass concentration at Molenbeek-Average Sunday and average working day during 'May-September 2009' and Car Free Sunday 20/09/2009

High PM10 values and a high PM2.5/PM10 mass to mass ratio have already been observed several times under comparable conditions, also on Sundays or official holidays with far less traffic as

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usual, a limited contribution of domestic heating and industrial activity and even in the presence of low concentrations for the gaseous pollutants. Common factors seemed to be a mild temperature (8 to 20°C) and a high humidity range (75 to 90%). As in the case of this car free Sunday, the elevated PM10 concentrations cannot be explained by a poor dispersion (IBGE technical report, 2009a). The average concentration computed for all eight car free Sundays (2002-2009) organized so far is of the same order or slightly higher than the average concentration on all Sundays or all working days. Figure 5 represents the evolution of the average PM10 concentration for all the Car Free Sundays, the average Sundays and the average working days in the different periods May–September 2002-2009. Unlike it is the case for the traffic related gaseous pollutants, one cannot observe a sharp or sudden change in the PM10 concentration neither at the beginning nor at the end of the traffic ban period, nor a clear sink of the concentration during traffic ban hours.

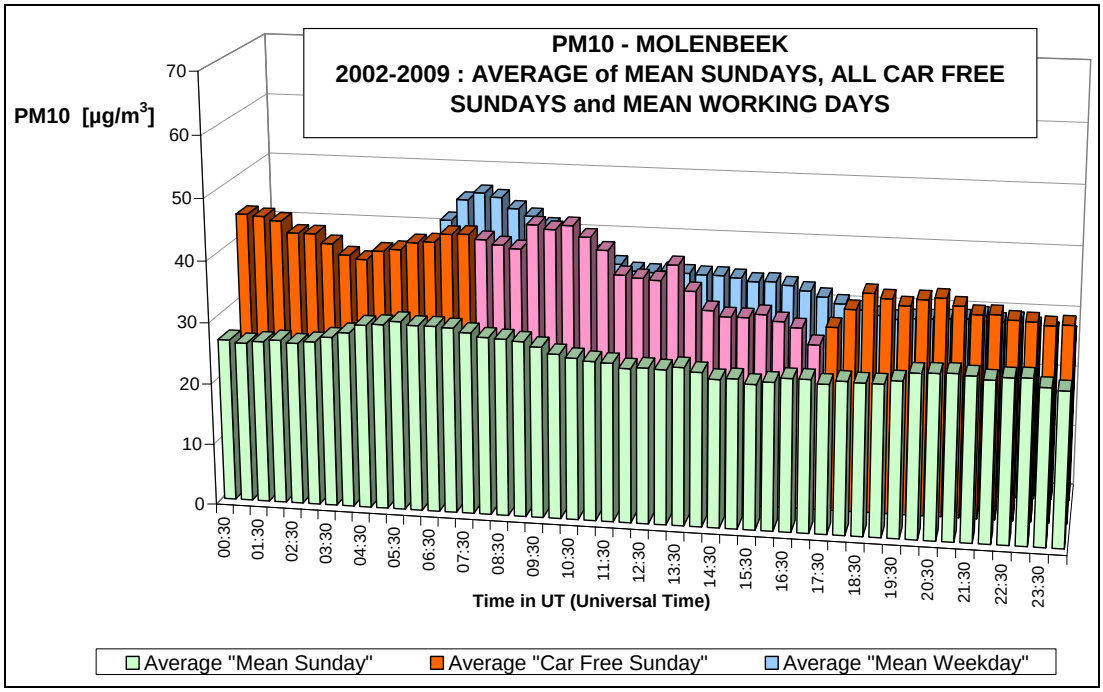

Fig. 5. PM10 mass concentration at Molenbeek - Average Sunday, average of all eight car free Sundays (2002-2009) and average working day during May – September 2002-2009

The evolution of the PM10 and PM2.5 concentrations was quite similar in all Brussels measuring sites, traffic as well as background stations (IBGE technical report, 2009b). At the Woluwe traffic site, the Black Carbon concentration however showed a continuous decrease during the traffic ban period, with a sharp decrease at the start and a sudden increase of the concentration as soon as the traffic returned. Figure 6 represents, for the period 18-22 September 2009, the half hourly values for PM10, NO and Black Carbon. During the traffic ban hours (centre of the graph) the evolution of the PM10 concentration is quite in opposition with that of Black Carbon, reflecting their different origin. The Black Carbon concentration does not always follow the PM10 pattern, rather it follows the pattern of the traffic related NO, as can be seen from the results on Saturday 19 and during the rush hours of Monday 21 September. Over a longer period, July 2009–June 2010, Black Carbon represents about 10% of the total PM10 mass concentration (IBGE technical report, 2010).

The TEOM-FDMS mass system also enables to register the loss of mass due to the presence of volatile or dissociating components, indicated as VO10 in the graph. The presence of a non-negligible quantity VO10 supports the idea that, as it was the case during the 2006 car free Sunday, the formation of secondary aerosol is responsible for the high PM mass concentration. Analysis of additional taken filters revealed that ammonium, nitrate and sulfate accounted for about 30% of the PM2.5 mass fraction. The mild temperature and high humidity are in favor of a stable ammonium nitrate aerosol (Stelson, 1982)

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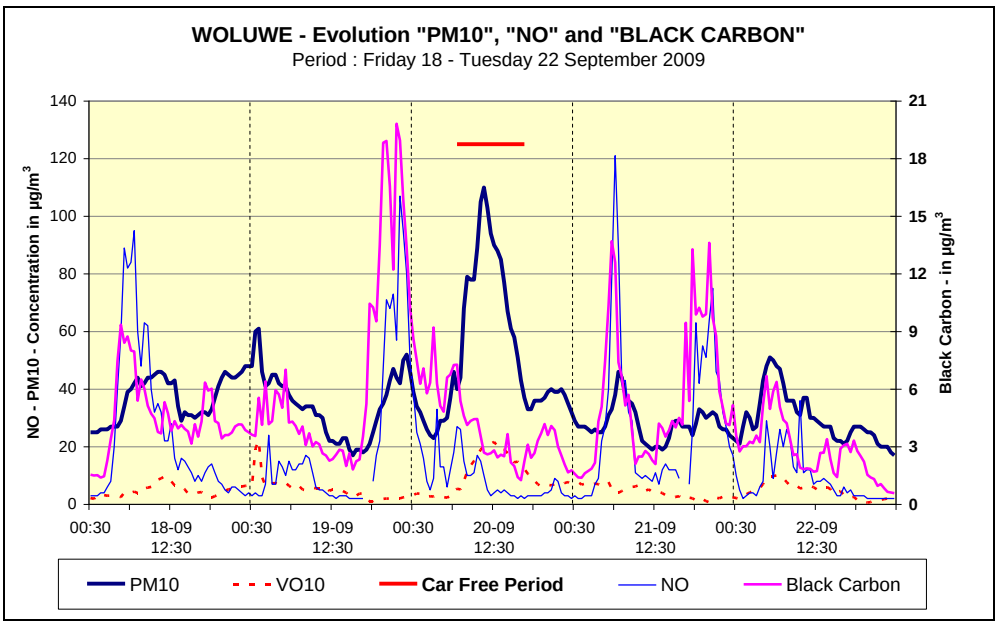

Fig. 6. Woluwe-Dynamic evolution for Black Smoke, NO, PM10 and the loss of volatile fraction (VO10) during sampling. Period Friday 18–Tuesday 22 September 2009

Particulate Number Concentration

At the Woluwe highway site, the number of particulates are counted and classified into 31 categories with equivalent diameters between 0.25 µm and 32 µm. For the classes with a diameter below 2.5 µm there is a striking increase of the particulate number concentration, especially during the traffic ban hours. For the smallest particulates, ranging between 0.25 and 0.28 µm, the increase of the numbers is most apparent at the beginning of the traffic ban, between 7:00 and 10:00 h UT (Fig. 7).

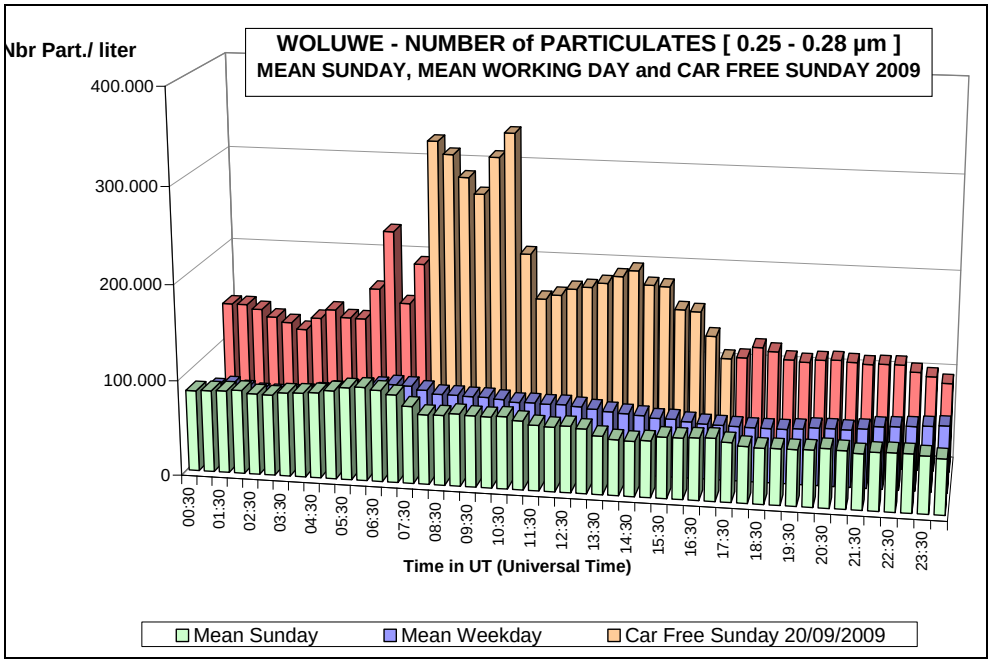

Fig. 7. Particulate Number concentration [0.25 – 0.28 µm] - Average Sunday and average working day during 'May-September 2009' and Car Free Sunday 20/09/2009

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For the particulates of a slightly larger size, with an equivalent diameter between 0.50 and 0.65 µm, the maximum number is reached only two hours later, between 10:00 and 12:00 h UT (Fig. 8). For a still larger particulate size, with diameter between 1.00 and 1.60 µm, the highest number concentration is observed between 10:30 and 13:00 h UT, collectively suggesting a time-dependent particulate growth process. The total number of all particulates greater than 0.25 µm reached a maximum of 1,200,000 (1.2 million) particulates per litre of ambient air between 9:30 and 10:00 h UT and represents one of the highest half hourly values (99.8^{the} percentile) of the year 2009 (IBGE technical report, 2010). High particulate numbers, above 1 million particulates (>0.25 µm) per litre air, are regularly observed during periods with formation of secondary aerosol. The record value since the start of the measurements end June 2008, is about 1,690,000 particulates per litre. This case proves, once again, that high particulate mass concentration, as well as high particulate number concentration may not automatically be associated with particulate emissions originating directly from traffic.

For the coarser fraction, particulates between 2.5 and 10 µm, the evolution of the particulate numbers of the 2009 car free Sunday follow that of an average Sunday. No peak value is observed during the interdiction hours (IBGE technical report, 2009b). In the absence of traffic and turbulences created by the traffic, there is less (re)suspension of the coarser particulates already deposited near or on the road surface.

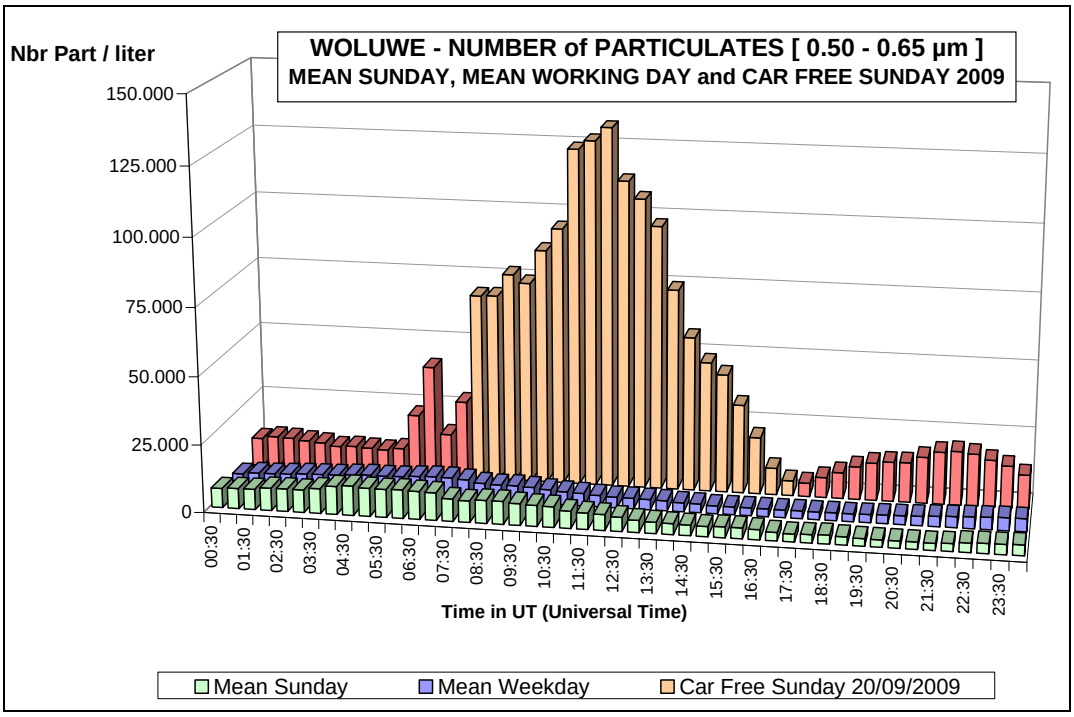

Fig. 8. Particulate Number concentration [0.50 – 0.65 µm] - Average Sunday and average working day during 'May-September 2009' and Car Free Sunday 20/09/2009

DISCUSSION AND CONCLUSION

For the organization of the different Car Free Sundays the Brussels Authorities achieved the maximum possible for banning the motorized traffic in the entire Brussels Capital Region. During the interdiction hours, the concentration of traffic related gaseous pollutants, mainly nitrogen oxides, decreased significantly in road tunnels. Generally this is also true for traffic oriented ambient measuring sites. Sharp and sudden concentration changes are normally observed at the beginning and at the end of the traffic ban hours. In the past, similar observations could be made for any of the car free Sundays, organized under different meteorological conditions. Averaged over all car free Sundays, the NO_2 concentration decreased at all measuring sites of the Brussels Capital Region, in the traffic stations as well as in the suburban and background stations. This observation makes clear that compliance with the EU limit value for the annual average concentration can and will be achieved, but only provided that further drastic NO_X traffic emission would be reduced.

During the car free Sunday of 20 September 2009, and especially during the traffic ban hours,

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very high PM10 and PM2.5 concentrations were observed in the whole Brussels urban area. During the interdiction hours and due to the importation of air masses polluted with secondary aerosol, PM concentrations levels were two times higher than those outside that period, when traffic was allowed. The absence of a sharp and sudden concentration change at the beginning and at the end of the traffic interdiction period seems to indicate that direct particle emissions from traffic only contribute to a slight extent in the total PM10 or PM2.5 mass concentration.

As it was the case for the 2006 car free Sunday, the daily average PM10 concentrations exceeded the EU limit value of 50 µg/m^3 in several PM10 measuring sites. This happened despite the absence of traffic during part of the day and a limited contribution of domestic heating and commercial or industrial activities. In both cases the formation of secondary aerosols was the main reason for the presence of these high PM concentrations. A strong indication for the non-negligible role of secondary aerosol is found in the fact that the PM2.5 to PM10 mass to mass ratio ranged as high as 80 to 90% and that the presence of volatile material, mainly inside the PM2.5 fraction was confirmed with about 30% of the total PM2.5 mass concentration identified as ammonium, sulfate and nitrate.

At the urban highway station the Black Carbon concentration seems well correlated with the traffic related NO concentration and hence to reflect the presence of traffic. Black Carbon is the only PM component showing a continuous concentration decrease during the traffic ban hours. A sudden concentration change was observed at the start of the traffic ban and a concentration increase as soon as the traffic returned. Averaged over a period of several months Black Carbon at this urban highway station represents about 10% of the total PM10 mass concentration.

Together with the PM10 and PM2.5 mass concentration, also the particle number concentration showed a sharp increase at the beginning of the traffic ban period. At its maximum, at 10:00 h UT, in the middle of the traffic ban hours, the total number of particulates with equivalent diameter above 0.25 µm reached one of the highest values of the year 2009. High number concentrations were obtained for all classes with diameters between 0.25 and 1.60 µm. For the coarser particulates, with diameter between 2.5 and 10 µm, no such increase was observed. The absence of turbulences created by the traffic put a limitation on the (re)suspension of that fraction.

Observations on car free Sundays lead to the conclusion that a traffic ban has an immediate and important beneficial effect on the concentration of traffic related gaseous pollutants and on the presence of Black Carbon in the ambient air, especially at traffic oriented measuring sites. A traffic ban has only a limited effect on the PM10 and PM2.5 mass concentration. The experience with the car free Sundays also helped to understand that the frequently occurring very high PM mass concentration are not necessarily caused by the particles directly emitted by the local traffic. Other phenomena, such as the presence of secondary formed aerosols may have a much greater contribution to the total particle mass and number concentration.

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