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Abstract : *On September 22nd 1998, 35 French towns took part in a pilot experiment consisting in drastically reducing the vehicle traffic in the town centres. According to that scheme, INERIS, in association with the local Air Quality Monitoring Network AIRLOR, carried out a BTX sampling campaign over 3 days, in order to quantify the impact of the traffic restrictions on air quality. This campaign was carried out in the centre of Nancy, where traffic restrictions were imposed on an extensive area. Sampling sites were chosen close to the busiest traffic routes. Sampling was carried out using passive samplers, on 10 hours on each day of the campaign. Measurements show clearly that, over the 3 days, the traffic flow reduction led to a decrease in BTX roadside concentrations by between 30 to 80 %. These measurements correlate well with CO levels monitored at one of the sampling sites, and with the volume of traffic in circulation each day. The correlation between BTX and CO clearly indicates traffic as the main source of atmospheric pollution. Measured benzene levels over the 3 days are compared with statutory limit values in France and in Europe.*

Key words *: urban atmospheric pollution, vehicular traffic reduction, BTX*

Résumé *: Le 22 septembre 1998, une trentaine de villes françaises ont participé à une journée pilote de réduction du trafic automobile en centre ville. L'INERIS s'est associé à cette opération avec le réseau AIRLOR, en procédant à des prélèvements de benzène-toluène-xylènes (BTX) afin de quantifier l'impact de la réduction du trafic sur les niveaux de pollution au centre ville de NANCY, où la zone de circulation réglementée était assez étendue. Les sites ont été choisis en raison de leur proximité avec le trafic. Les prélèvements ont été effectués à l'aide de tubes passifs pendant une dizaine d'heures lors de la journée sans voitures, ainsi que la veille et le lendemain. Les mesures montrent clairement que la réduction du trafic automobile induit une réduction des concentrations en BTX dans les sites de proximité. La diminution des concentrations varie selon les sites entre 30 % et 80 %, voire plus pour les composés les plus lourds. Ces diminutions de concentrations sont bien corrélées avec les autres données fournies par AIRLOR, à savoir les teneurs en CO mesuré en l'un des points, et le nombre de véhicules relevé par deux boucles de comptage de trafic. La corrélation entre BTX et CO est typique d'une pollution issue principalement du trafic automobile. La valeur des concentrations en benzène mesurées en présence ou en l'absence de voitures, est comparée aux valeurs réglementaires en France et en Europe.*

Mots-clés *: pollution atmosphérique urbaine, réduction de la circulation automobile, BTX*

Introduction

In 1997, the City of La Rochelle, concerned with urban pollution problems, decided to restrict the flow of traffic in its town centre for one day. This operation is now repeated on an annual basis each 22nd of September, with an increasing number of French towns involved (35 in 1998, more than 60 in 1999). This operation is managed by the French Ministry of Environment.

The aims of this operation are « to give the town back to pedestrians and cyclists, to develop public transport facilities, and to improve the air quality in town centres. » (Press release of the French Ministry of Environment, 1998).

1. Objectives

It seems obvious that with limited traffic or even no traffic at all, travelling is easier for pedestrians, cyclists and buses. However, assessment of any real improvement in air quality has to be carefully carried out by appropriate means. Therefore, in the different participating towns, the local Air Quality Monitoring Networks have set up additional monitoring stations. The role of INERIS was to quantify the reduction of pollution related to vehicular traffic by measuring BTX (benzene, toluene, xylenes) levels. The sampling campaign was carried out in collaboration with AIRLOR (the Air Quality Monitoring Network for the Lorraine region, France), with whom a previous study of personal, indoor and outdoor exposure to VOC had been carried out (Gonzalez-Flesca et al, 1999)

Samples were taken over three days, starting the day before (September 21st, identified as day "A") and ending the day after (day "C"), the "traffic-free" day (day "B"), in order to evaluate the changes in BTX levels due to the restriction of traffic, and taking into account the influence of meteorological variations.

The results of the BTX analysis could then be completed with the following data supplied by AIRLOR : CO concentrations at different points in the town centre, and volume of traffic circulating in the centre during those days.

2. Experimental

Out of the 15 sites reviewed, the 10 retained were chosen because they came forward as good candidates for high roadside pollution levels : crossroads or busy streets, roads where the traffic was slowed down due to corners or bends... One of the sites was situated beside an AIRLOR monitoring station.

Each day, sampling was carried out using axial diffusion tubes loaded with "Carbotrap B", previously described (Bates et al, 1997). The tubes were placed inside purpose built aluminium shelters to prevent vandalism. Thermal desorption followed by GC analysis of the tubes was performed at INERIS.

3. Results and discussion

Comparison of BTX concentrations over the three days

BTX concentrations have been determined for all sites. Results are shown in Table 1.

Day A, site n°	1	2	3	4	5	6	7	8	9	10
BENZENE	7.4	3.4	3.1	4.2	3.6	2.4	4.7	2.5	2.4	2.3
TOLUENE	15.9	4.7	6.2	8.7	6.9	6.0	6.2	4.4	4.3	2.6
m+p-XYLENE	11.7	3.7	4.6	6.5	5.0	3.6	4.5	3.2	4.6	1.5
o-XYLENE	4.0	1.4	1.6	2.4	1.7	1.2	1.8	1.5	1.1	0.6

Day B, site n°	1	2	3	4	5	6	7	8	9	10
BENZENE	2.7	0.8	2.1	1.0	2.3	0.7	2.0	1.1	1.3	1.4
TOLUENE	2.9	1.3	2.7	1.7	1.7	1.6	3.2	1.1	1.2	1.7
m+p-XYLENE	2.0	1.1	1.4	0.5	1.4	1.0	1.1	0.8	0.9	1.1
o-XYLENE	0.8	0.0	0.6	0.0	0.6	0.0	0.7	0.6	0.5	0.3

Day C, site n°	1	2	3	4	5	6	7	8	9	10
BENZENE	8.0	3.5	6.0	6.7	6.1	5.3	6.9	3.7	5.6	3.0
TOLUENE	20.2	7.3	8.6	15.4	14.1	12.2	16.9	9.0	13.1	5.5
m+p-XYLENE	16.2	5.1	5.9	11.7	9.9	8.6	11.6	6.5	9.6	4.0
o-XYLENE	5.6	1.7	2.5	4.1	3.6	3.2	4.1	2.5	3.6	1.6

Table 1 : BTX concentrations in $\mu g/m^3$ for each site, for each day of the campaign

Tableau 1 : concentrations des BTX en $\mu g/m^3$ par site, pour chacun des 3 jours

The wind direction : E-NE, was steady over the three days, whereas the mean wind speed was higher on days A and B : around 4 m/s, than on day C : below 2 m/s, resulting in a greater dispersion of pollutants. In order to compare the concentrations measured over the three days, we have plotted "normalised" values for each compound, expressed as {[concentration, $\mu g/m^3$]×wind speed}. These results are shown in Figures 1 to 4.

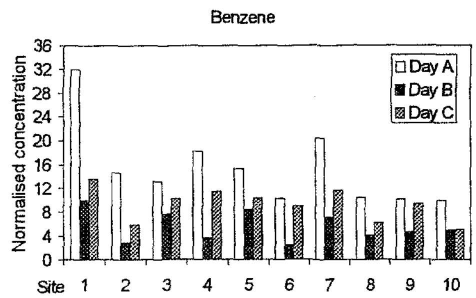

Figure 1 : normalised benzene levels over the 3 days
Figure 1 : concentrations normalisées en benzène

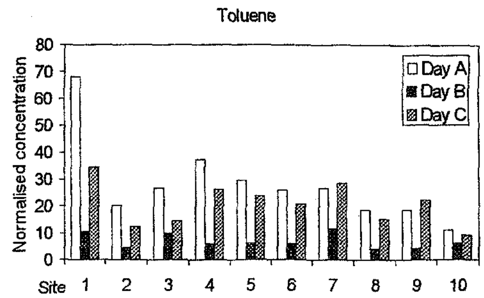

Figure 2 : normalised toluene levels over the 3 days
Figure 2 : concentrations normalisées en toluène

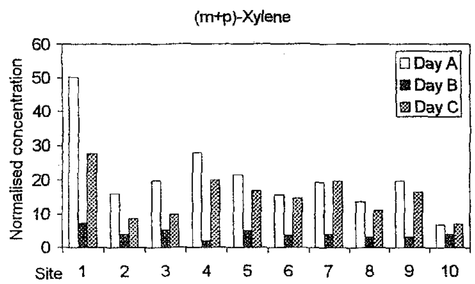

Figure 3 : normalised (m+p)xylene levels over the 3 days
Figure 3 : concentrations normalisées en (m+p)xylène

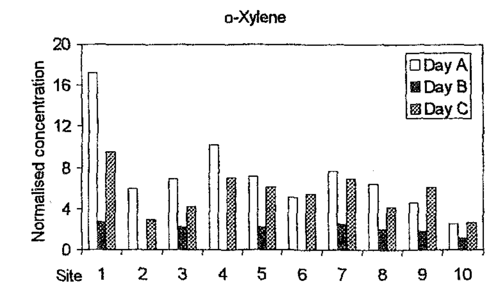

Figure 4 : normalised o-xylene levels over the 3 days
Figure 4 : concentrations normalisées en o-xylène

Figures 1 to 4 show clearly the decrease of roadside BTX concentrations due to the reduction in the volume of the traffic, except for site n° 10 where the values are low : this site is more representative of background concentrations, which are far less affected by the limitation of traffic.

It should be noted that sampling site n° 1, which was situated beside the AIRLOR monitoring station "Henri Poincaré", always exhibited the highest BTX concentrations. At this site, wind direction was parallel to the street axis over the three days.

BTX concentrations for day B have been compared with those of days A and C. The decrease in concentrations has been calculated as follows :

$$d(\%) = 1 - \left(\frac{C_B^i}{C_{A,C}^i} \right)_s \times 100$$

Where C_B^i = concentration, in $\mu g/m^3$, of the compound **i**, measured in the site **s**, on day **B**, and $C_{A,C}^i$ = idem but on days A or C.

The reduction in concentration, **d**, varied between 30 to 80 %, depending on the site and compound, as shown in Table 2. The greatest differences were observed for the heavier compounds. For some sites, o-xylene concentrations on the « traffic-free » day were below the GC quantification limit : this gives an apparent "100 %" decrease.

Decrease of concentration related to the day before, %

Site n°	1	2	3	4	5	6	7	8	9	10
BENZENE	64	77	32	76	36	72	58	55	46	41
TOLUENE	82	73	56	81	76	73	48	74	73	35
m+p-XYLENE	83	71	69	92	73	72	76	74	81	29
o-XYLENE	81	100*	62	100*	64	100*	61	62	53	46

Decrease of concentration related to the day after, %

Site n°	1	2	3	4	5	6	7	8	9	10
BENZENE	66	78	65	85	63	87	72	70	77	55
TOLUENE	86	83	68	89	88	87	81	87	91	69
m+p-XYLENE	88	79	76	95	86	88	91	87	91	73
o-XYLENE	87	100*	76	100*	83	100*	83	77	86	80

** : for these samples the concentration on "trafic-free" day was below quantification limits.*

Table 2 : Concentration ratios between the days « with » and « without » cars
Tableau 2 : rapports de concentrations en BTX entre les jours « avec » et « sans » voitures

Correlation of BTX concentrations with other data

Carbon monoxide concentrations over 4 days are reported in Figure 5, volume of traffic in Table 3 (Bourdet, 1998). The decrease in BTX concentrations in the traffic restriction zone of the centre of Nancy, correlates well with CO levels monitored by AIRLOR at site n°1, and with the volume of traffic recorded at two points, in Lafayette Street (between sites n° 6 and 8) and Stanislas Street (near site n° 5).

Though data are missing between 9 a.m. and 2 p.m. (7 to 12 a.m, GMT) due to a monitoring failure, it can be seen that CO concentrations recorded on the « traffic-free » day does not show the morning rush hour peak, as the traffic was restricted from 7 am (5 a.m, GMT as indicated on Fig. 5). The sudden increase in concentration at 5 p.m. GMT (7 p.m. local time) corresponds to the end of the restriction : we saw the traffic returning to normal within a few minutes after 7 p.m. Nevertheless, the integrated CO concentration for this day is, according to AIRLOR, 5 to 7 times smaller than usual. Likewise, the decrease in BTX concentrations, by 60 to 70 %, is comparable to the trends in the volume of traffic. Correlation between BTX concentrations and CO levels are typical of roadside pollution (Gonzalez-Flesca et al, 1998).

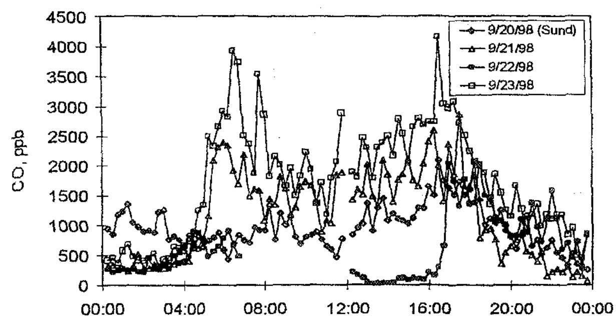

Figure 5 : CO concentrations at site N°1 ("Poincaré") during the sampling days
Fig. 5 : profils de CO à la station POINCARE (site n°1) lors des jours de prélèvements

Counting site	Total number of vehicles/day				Decrease Tuesday/ Monday
	09/19/1998	09/20/1998	09/21/1998	09/22/1998	
Stanislas St	17 801	12 963	17 747	5 751	-67 %
Saint-Jean St*	815	352	1 060	947	-10 %
Lafayette St	4 146	3 650	3 667	1 275	-65 %

* Saint-Jean Street : this street is a 2-lane road limited to bus traffic, this one normal on Tuesday 22nd.
Rue St Jean : *cette rue ne comporte que deux voies de circulation réservées aux autobus, qui circulaient normalement le 22 septembre 1998*

Table 3 : Number of vehicles (Bourdet, 1998)
Tableau 3 : comptage des véhicules

BTX ratios

The daily ratios of toluene and xylenes over benzene were calculated for each site : the values are shown in Table 4.

Toluene/benzene ratio for the sampling sites

Date	1	2	3	4	5	6	7	8	9	10	Mean	St. deviation
09/21/1998	2.1	1.4	2.0	2.1	1.9	2.5	1.3	1.8	1.8	1.1	1.8	0.4
09/22/1998	1.1	1.6	1.3	1.7	0.7	2.4	1.6	1.0	0.9	1.2	1.4	0.5
09/23/1998	2.5	2.1	1.4	2.3	2.3	2.3	2.4	2.4	2.3	1.8	2.2	0.3

(m+p)-Xylene/benzene ratio for the sampling sites

Date	1	2	3	4	5	6	7	8	9	10	Mean	St. deviation
09/21/1998	1.6	1.1	1.5	1.5	1.4	1.5	0.9	1.3	1.9	0.7	1.3	0.4
09/22/1998	0.7	1.4	0.7	0.5	0.6	1.5	0.5	0.7	0.7	0.8	0.8	0.3
09/23/1998	2.0	1.4	1.0	1.7	1.6	1.6	1.7	1.8	1.7	1.3	1.6	0.3

o-Xylene/benzene ratio for the sampling sites

Date	1	2	3	4	5	6	7	8	9	10	Mean	St. deviation
09/21/1998	0.5	0.4	0.5	0.6	0.5	0.5	0.4	0.6	0.4	0.3	0.5	0.1
09/22/1998	0.3	**	0.3	**	0.3	**	0.4	0.5	0.4	0.2	0.3	0.1
09/23/1998	0.7	0.5	0.4	0.6	0.6	0.6	0.6	0.7	0.6	0.5	0.6	0.1

*** : no ratio available, o-xylene concentration was below quantification limits*

Table 4 : BTX ratios for the different sampling days
Tableau 4 : rapports des BTX entre eux, selon le jour de prélèvement

The ratio of toluene/benzene has a mean value of 2 for the days having normal traffic and 1.4 for the « traffic-free » day. Likewise, benzene/xylenes ratios show a decrease in the heavier compounds on the day with restricted traffic. This is typical of an aged air mass, when the major pollution source is the traffic. Oxidation of hydrocarbons in the troposphere involves mainly $^{\bullet}OH$ during the day, and $NO^{\bullet}_3$ at night. Reactivities of oxidation vary from one hydrocarbon to another, in the following sequence :

m-xylene > (o, p)-xylene > toluene > benzene (Académie des Sciences, 1993)

Near a source like a busy street, the benzene/toluene ratio will be given directly by the source itself, e. g. fuel and exhaust composition. During the « traffic-free » day, when the direct source was limited, the BTX levels came from either older or remote emissions. In this way, tropospheric oxidation could occur, and cover the local diffusion or dispersion effects. Therefore, the BTX ratios are modified according to reactivities.

Benzene levels

Nearly all the roadside benzene levels measured at the 10 sites in the centre of Nancy, under light wind conditions, exceed the EC proposed limit value of 5 $\mu g/m^3$ (Proposed Directive COM 1998, 591). All of them exceed the French target value for air quality of 2 $\mu g/m^3$ (Décret Français n° 98-360, 1998). With traffic restrictions, all the sites show a benzene concentration below 5 $\mu g/m^3$, and nearly all the sites are close to the French target for air quality, the greatest value being 2.7 $\mu g/m^3$.

Conclusion

This roadside BTX study carried out in the centre of Nancy shows that passive sampling is a useful method for measuring hydrocarbons even for periods as short as 10 hours. Direct comparison between pollution levels at different traffic volumes is possible, provided little changes in the meteorological conditions are involved. We have shown a good correlation between BTX concentrations, CO levels and the volume of traffic over the 3 days of the campaign, which is typical of a roadside pollution.

Traffic restrictions greatly improve the air quality, especially under light wind conditions : with a normal flow of traffic, roadside benzene levels are always higher than the French Target value for Air Quality, of 2 $\mu g/m^3$, and exceeds, in most places, the proposed European Limit Value of 5 $\mu g/m^3$. With a limited flow of traffic, all sites show a benzene concentration smaller than the European Limit Value, and the French Target value is only slightly exceeded.

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