Air Pollution Measurements in Traffic Tunnels

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Air pollution measurements during April 1991 are reported from the Craeybeckx highway tunnel in Antwerp, Belgium. The tunnel was used daily by an average of 49,000 vehicles, of which 60% were gasoline fueled passenger cars, 20% diesel cars, and 20% trucks. Of the gasoline cars, only 3% had three-way catalytic systems. Tunnel air concentrations of nitrogen oxides, sulphur dioxide, carbon monoxide, nonmethane hydrocarbons, volatile organic compounds, polycyclic aromatic hydrocarbons, and lead are presented. The traffic emissions in the tunnel are calculated by the carbon balance method, which uses the increase of the total carbon concentration in the tunnel air as the reference quantity. Division of the concentration of any pollutant by the total carbon concentration gives emission factors per kilogram of carbon. These emission factors can be converted directly to emissions relative to fuel consumption or per kilometer. The fraction of diesel used in the tunnel was derived from sulphur to carbon ratios in tunnel air. A calculation procedure with breakdown of emission factors according to vehicle categories was used to estimate countrywide emissions. The estimated emissions were compared to results from the Flanders Emissions Inventory (Emissie Inventaris Vlaamse Regio [EIVR]) and calculated emissions according to the emission factors proposed by the European Commissions CORINAIR Working Group. For NOx there is excellent agreement. For carbon monoxide and hydrocarbons, the tunnel data produced higher emissions than the CORINAIR model would predict but lower than the official EIVR statistics. The estimated lead emissions from traffic are found to be 22 to 29% of the lead in gasoline. — Environ Health Perspect 102(Suppl 4):31–37 (1994).

Key words: air pollution, traffic tunnel, emission factors, carbon balance, corinair

Introduction

Air quality in traffic tunnels has received much attention in Belgium since 1989 when the Institute for Hygiene and Epidemiology (IHE), after measurements in two traffic tunnels in Brussels, published rather alarming results. It appeared that (a driver can get caught in stagnant traffic and be obliged to remain 20 min or more in a severely polluted atmosphere (e.g., in the 2-km long Leopold II tunnel). Under these circumstances or for frequent tunnel users, it was shown that the exposure limits set by ambient air quality standards are exceeded. Since then, the air quality in most of the larger tunnels in Belgium has been investigated by the IHE. As a result of their work, Vanderstraeten et al. (1) developed the carbon balance method to estimate on the road traffic emissions from air pollution measurements in a tunnel. The method can be regarded as an extension of the tracer method described by Bullin et al. (2) with the carbon from the fuel used as a tracer for the exhaust gases.

In this paper, results for air pollution measurements by the Flemish Institute for Technological Research in the Antwerp Craeybeckx tunnel in 1991, and an application of the carbon balance method to estimate emissions of the major pollutants is presented. As opposed to Brussels’ city tunnels where only passenger cars are allowed and the speed is limited to 60 to 80 km/hr, this is a highway tunnel with high speed traffic and intensive freight traffic (20% trucks).

The Craeybeckx Tunnel

The Craeybeckx tunnel (Figure 1) is a 1600-m long highway tunnel with two tubes having five lanes in each direction, one of which is reserved for emergency parking. The south direction of the tunnel is the highway to Brussels, and in the north, it is connected to the Antwerp urban area. Because it is the highway link between the largest Belgian cities, the tunnel is used intensively by all kinds of motor traffic. The tunnel has a very smooth curvature to the west and negligible slopes. During most of the day, there is a typical highway traffic flow with average speeds from 90 to 120 km/hr. The tunnel galleries are 6 m high by 21 m wide and are separated by a central service alley containing 33 air exhausts. Fresh air can be blown in by 33 regularly spaced ports in each of the outside walls. The tunnel ventilation is activated when carbon monoxide (CO) levels exceed 50 ppm, but because this seldom occurs, it has become practice to activate part of the blowers for a fixed time during peak hours. The site of the measurements was chosen at one third from the end in the northbound tube because this direction was most sensitive to congestion in morning and evening rush hours and thus was expected to offer a variety in traffic and emission patterns.

The number of vehicles passing through the northbound tube was 45,000/day (working days) and 23,000/day (weekends), with hourly maxima around 5000 and 4000 for morning and evening peaks, respectively.

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Figure 1. The Craeybeckx tunnel. A, point of measurements in tunnel; B, point of background measurements.
The fraction of trucks was counted as 21% on a per day basis and as 18% during rush hours from 7:30 to 9:00 A.M. and 4:30 to 6:30 P.M. For emission calculations, a breakdown into 20% diesel passenger cars, 20% trucks, and 60% gasoline cars is assumed, with each class subdivided into power and weight classes according to the national statistics. It is important to note that only 3% of the gasoline cars had three-way catalysts during the measurements.

**Experimental**

Air pollution measurements were carried out from March to April 1991 by both continuous monitoring equipment and integrated samplers. The complete experimental apparatus is described in the report by Bruynseraede (3). Nitrogen oxides were analyzed by chemiluminescence with catalytic nitrogen dioxide (NO₂) converter, nonmethane hydrocarbons (NMHC) by flame ionization detection, sulfur dioxide by UV fluorescence, and carbon monoxide and carbon dioxide (CO₂) by nondispersive infrared analyzers. Wind speed in the tunnel was measured by a Young cup anemometer mounted at a height of 2.5 m and 2 m from the tunnel wall. The data were stored on computer disk as 5-min averages. For a number of days, car speeds and counts per lane were registered from the tunnel’s electronic system.

Volatile organic compounds (VOCs) were sampled as 2- and 4-hr averages in air sample bags and on activated charcoal. Particulate matter, including polycyclic aromatic hydrocarbons (PAHs), was collected by high-volume samplers on glass fiber filter followed by polyurethane foam absorbent as 24-hr averages. Lead particles and asbestos fibres were sampled occasionally on membrane filters as 24-hr average by medium volume sampling at a rate of 1 m³/hr.

**Background Concentrations**

The importance of accurate information on the pollution levels in the air entering the tunnel will be demonstrated further. Preferably these levels should be monitored at the air intake of the tunnel and simultaneously with the tunnel measurements. For practical reasons, this was not feasible for all components. Only sulfur dioxide (SO₂) and CO₂ were monitored simultaneously with a second analyzer on top of the tunnel. Nitrogen oxide (NO₂) data were obtained from local national air pollution network stations and CO background levels appeared to be virtually zero. VOC and particulate measurements were carried out on separate days with the equipment that was used for the tunnel measurements.

**Air Pollution Data**

Figure 2 shows air concentration histories of some of the pollutants monitored during an 11-day period from April 17 (Wednesday) to 27 (Saturday) 1991. On working days, peak concentrations of all pollutants occur in the morning from 7 to 9 A.M. and in the evening from 5 to 7 P.M. It is obvious that the pollution levels in the tunnel are determined primarily by the traffic intensity, of which the pattern is shown in Figure 3. Because this tunnel has an impressive cross section and efficient ventilation, the pollutant concentrations are lower by a factor of 2 or more than in other Belgian tunnels of comparable length. The main statistics for the half-hour averages represented in Figure 2 are shown in Table 1.

The data refer to an ambient air temperature of 10°C and a pressure of 1013 mbar. Analysis of the air pollution data in combination with vehicle speed indicates that CO, hydrocarbon and NO₂ emissions increase markedly when traffic becomes congested. Average data for some of the VOCs during day time, and a selection of PAHs together with standard deviations and background levels are given in Table 2.

The VOC data are averages of 29 samples taken on 14 days, covering different periods. The large standard deviations are mainly due to variance between these periods. The PAH data are an average of nine samples of 24-hr averages. The PAH profile, obtained by ratioing to benzo[α]pyrene concentrations is remarkably similar to those from six tunnel studies cited by Daisey (4). With the exception of pyrene, chrysene, and anthanthrene our data have the same profile as Grimmer’s 1980 data obtained in Essen, Germany.

**Relation to Air Quality Standards**

Most of the measurements of tunnel air in Belgium so far were initiated after complaints from tunnel users. Proper standards for tunnel air quality are not yet available, however. If the tunnel air quality were to be compared to Belgian ambient air quality standards (3), there would be an exceedance of the NO₂ standard of 200 μg/m³ as the 98 percentile of hourly values and of the SO₂ standard of 80 μg/m³ as the median of daily values. The data further indicate that probably the black smoke standard of 80 μg/m³ as the median of daily values is exceeded and that lead
remains below the 2 μg/m³ daily average limit (Table 3). For the other pollutants like nitric oxide (NO), VOCs, and PAHs, there are currently no legal air quality standards.

However, from the definition of their observation times alone, it is understood that ambient air standards cannot apply to tunnels. To ensure air quality, the Craeybeckx tunnel authorities operate a CO monitoring system to control the ventilation in three segments of the tunnel. The CO threshold to start ventilation in a tunnel segment is 50 ppm. This method of operation is well in line with the 1987 WHO Air Quality Guidelines for Europe (5), where a 50 ppm CO limit is given for a 30-min exposure time. For most pollutants, however, no short time exposure levels for the general public are available. The development of practical tunnel air quality standards that would take into consideration the residence time of the tunnel users and the maximum allowable exposure to the most critical pollutants: NO₂, SO₂, lead, and particles like benzene and certain PAHs, is highly recommended.

**Mass Balances and Carbon Balance**

Air pollution measurements in combination with mass balances over a traffic tunnel allowed a calculation of the average emissions from the vehicles using the tunnel. The parameters measured were a) air flow or average wind speed over the tunnel section, b) concentration of pollutants of interest in the air entering and leaving the tunnel (in the following, the difference between these concentrations is named the net concentration in the tunnel), and c) the number of cars passing through the tunnel, if the emissions are to be estimated per vehicle or per kilometer. In its simplest form, for a tunnel with longitudinal airflow, the mass balance result for emission of pollutant \( i \) is given by:

\[
E_i = \frac{\Delta y \times V \times S}{L \times n}
\]

### Table 1. Statistics of half-hour average concentrations in tunnel air.

| Compound | NO | NO₂ | NOₓ | CO | CO₂ | NMHC | SO₂ |
|----------|----|-----|-----|----|-----|------|-----|
| Average  | 1557 | 206 | 2593 | 3.33 | 979 | 1.04 | 122 |
| Maximum  | 265  | 657 | 8823 | 26.4 | 1742 | 4.82 | 224 |
| Minimum  | 271  | 63  | 507  | 0.5 | 744 | D.L.(<0.4) | 49 |

NMHC, nonmethane hydrocarbons.

**Figure 3.** Daily patterns of carbon monoxide (CO), nitric oxide (NO), and carbon dioxide (CO₂) concentrations in tunnel air. Tunnel wind speeds dominate the ventilation rate as is illustrated by CO₂ concentration. The average vehicle speed (bottom graph) determines CO and NO profiles.

**Table 2.** Average volatile organic compounds and poly-cyclic aromatic hydrocarbon concentrations in tunnel air.

| Concentration, μg/m³ | Tunnel | Background |
|-----------------------|--------|------------|
| Methane               | 1021 ± 49 | 1075 |
| Ethane                | 166 ± 58  | 30 |
| Propane               | 26 ± 11   | 5.8 |
| Isobutane             | 17 ± 5    | 6.6 |
| 1-Butene              | 13 ± 3.8  | 1.5 |
| Butane                | 24 ± 10   | 4.1 |
| Benzene               | 23 ± 9.6  | 1.3 |
| Toluene               | 58 ± 24   | 4.2 |
| Ethylbenzene          | 12 ± 5.7  | 1.2 |
| m- and p-Xylene       | 34 ± 14   | 2.4 |
| α-Xylene              | 17 ± 7    | 1.0 |
| 3-Ethyltoluene        | 23 ± 4    | 1.3 |
| 1,2,4-Trimethylenzene | 15 ± 7    | 1.0 |
| Decane                | 13 ± 21   | 0.1 |
| 1,2,3-Trimethylenzene | 4.7 ± 3.7 | 0.0 |
| Dodecane              | 2.9 ± 2   | 0.0 |
| Pentadecane           | 1.9 ± 0.8 | 0.2 |
| Pyrene                | 16 ± 3.5  | 0.9 |
| Chrysene              | 9.5 ± 2.1 | 0.4 |
| Benzo[a]anthracene    | 12 ± 2.9  | 0.1 |
| Benzo[fluoranthene]   | 12 ± 3.2  | 0.2 |
| Benzo[k]fluoranthene  | 9 ± 2.3   | 0.1 |
| Benzo[a]pyrene        | 2.8 ± 0.5 | 0.04 |
| Benzo[pyrene]         | 9.6 ± 2.8 | 0.1 |
| Benzo[glycine]        | 9.6 ± 2.6 | 0.1 |
| Perylene              | 1.5 ± 0.4 | 0.04 |
| Indeno[1,2,3,c,d]pyrene| 9.4 ± 3   | 0.3 |
| Dibenzo[a,c]anthracene| 1 ± 0.3   | 0.03 |
| pDibenzo[a,c]anthracene| 1 ± 0.3   | 0.03 |
| Benzo[g,h,i]pyrene    | 19 ± 6    | 0.4 |
| Anthracene            | 15 ± 0.3  | 0.01 |
| Coronene              | 7.5 ± 1.6 | 0.4 |

**AIR POLLUTION MEASUREMENTS IN TRAFFIC TUNNELS**
where $F_i = \text{average emission of pollutant } i \text{ (g/km)}$; $\Delta y_i = \text{increase of concentration of component } i \text{ in tunnel air (g/m$^3$)}$; $V_w = \text{average wind speed in tunnel (m/sec)}$; $n = \text{number of cars passing through the tunnel (sec$^{-1}$)}$; $L = \text{length of the tunnel (km)}$; $S = \text{section of the tunnel (m$^2$)}$. The approach of Equation 1 was used by several authors (6–8) to calculate traffic emissions in tunnels.

For the Craeybeckx tunnel the respective values of the constants in Equation 1 are $S = 128 \text{ m}^2$, $L = 1.6 \text{ km}$, $n = 0.54$ to 0.23 as a daily average, and $V_w = 2$ to 7 m/sec. The product $V_w \times S$ is the air flow through the tunnel and $L \times n$ is the total number of kilometers driven per second inside the tunnel. The mass balance can be made over a part of the tunnel (e.g., when air is analyzed in the tunnel [as in this case, with $L \times [1 \text{.0 km}])$ or from ventilation exhausts.

In many tunnels, the air flow is not constant because air is blown in or withdrawn at different locations. $V_w$ is the average wind speed over the entire cross section and therefore is difficult to determine experimentally. A large number of measuring points distributed over the tunnel section would be required, which is impeded by the traffic. In the Craeybeckx tunnel, the air drag of the vehicles and the natural wind dominate the ventilation. The wind direction during our observations was always the same as the traffic. No additional dilution effect from the forced ventilation could be measured. A remarkable decrease in tunnel ventilation with peaking pollution levels was measured on April 22, 1991, when an incidental traffic slowdown coincided with strong winds from the opposite direction reducing tunnel wind speeds to 1 m/sec. This incident is illustrated in Figure 3 and shows that air flow through the tunnel is not always a reliable parameter to calculate mass balances.

To take into account the effect of variable dilution of exhaust gases in the calculation of the mass balance, a tracer gas like sulfur hexafluoride, released by vehicles was used by Bullin et al. (2) and Hlavinka et al. (9). This tracer method can be applied to tunnels and open roads, but the representative dispersion of the tracer is hardly feasible for longer periods of time.

Carbon balance as a method to evaluate traffic emissions in situ was applied by Vanderstraeten et al. (1) for two tunnels in Brussels. The method does not rely on determination of the air flow but requires measurements of the increase of air concentrations of all carbon-containing pollutants in order to relate emissions to the amount of carbon produced by fuel combustion in the tunnel. Carbon balance is a mass balance method with the carbon content of the air used as the tracer gas. The first step in drawing a carbon balance is to make a table of net pollutants concentrations in the tunnel air $\Delta y_i$, and the next step is to calculate the contribution of each pollutant $i$ to the unit mass of carbon emitted in the tunnel. Finally, a table of $P_i$ values is obtained:

$$P_i = \frac{1000 \times \Delta y_i}{\sum x_i \times \Delta y_i} \quad [2]$$

where $P_i = \text{net mass of pollutant } i \text{ in tunnel atmosphere per net mass of carbon in tunnel atmosphere (g/kg)}$ and $x_i$ carbon content of pollutant $i$ (mass fraction).

It is obvious that $P_i$ is independent of the dilution and thus the determination of $P_i$ in tunnel air is equivalent to its direct measurement in the emissions. The relation to the average emission factor in the tunnel, $E_i$, in g/km, is: $E_i = P_i \times T$ where $T$ is the average fuel consumption in the tunnel, expressed in kilogram carbon per kilometer. From Equation 1, we can estimate the fuel consumption $T$ in the Craeybeckx tunnel at 0.06 kg C/km on working days and 0.04 kg C/km in weekends. The $\epsilon$ values calculated in this way suffer from the imprecision in the measurement of $V_w$, and further are too low by an estimated 20 to 30% because tunnel air is vented through the ventilation system, even when this is not activated.

When $\epsilon$ is calculated from fuel consumption data (10) and vehicle and speed distributions, $\epsilon = 0.090$ is obtained, with 52% of the carbon originating from diesel.

Application of Equations 2 and 3 is attractive because it immediately results in emissions given in g/km, for traffic conditions as in the tunnel. Countrywide emissions can be estimated by multiplication of $E_i$ with the annual 60 billion km driven in Belgium. More sophisticated computations were used to calculate national emissions from true vehicle and speed distributions. The headlines of this approach are given in the following paragraphs.

Because CO$_2$ and CO account for nearly 99% of the carbon emission in this tunnel, it is not crucial that all pollutants are taken into account for the results of the carbon balance. Essentially, a CO$_2$ measurement alone will already give a good approximation for the totality of carbon in the tunnel air. Furthermore, the carbon balance must not be limited to carbon containing molecules. It is equally convenient to include NO and SO$_2$ and eventually others in the carbon table. Carbon balances can be calculated for any period of time; however, we find it most convenient to integrate over the sampling periods of a full day (24 hr) and 2 and 4 hr, where the latter were defined as: night from 12 to 4 A.M., morning rush from 6 to 10 A.M., mid day from 12 to 4 P.M., and evening rush from 4:30 to 6:30 P.M.

**Statistical Treatment of Data**

Our measurements of tunnel air pollution, when used for estimation of emissions, can be classified according to traffic mode. The traffic in the tunnel between rush hours is typical of highways in Belgium. Therefore, data over these periods can be considered representative for highway emissions. The rush hours with congested traffic are assumed to approximate urban driving conditions. On the other hand measurement itself determines the periods over which average data are available. Instruments like those for continuous monitoring of CO, hydrocarbons, SO$_2$, and NO$_x$ allow integration over any time span, while VOCs were sampled and required 2 or 4 hr averaging times. Particulate matter and related pollutants including PAHs, heavy metals, and dioxins were measured as 24-hr averages.

The subdivision of measurements in data sets for periods of 2, 4, or 24 hr according to traffic mode allowed the calculation of four carbon balances for every day. A total of 22 data sets were of sufficient experimental quality. The hypothesis that there were statistical differences between those sets was tested by Kruskal Wallis analysis. This test is comparable to the better known F test, but it does not require normal distribution of the data. The four modes we defined initially were: a) nights with low-traffic intensity, b) normal traffic during morning and afternoons, c) intensive traffic during rush hours, and d) congested traffic during evening and morning jams. One result the test indicated with 90% or higher confidence that specific emissions of toluene and CO differ by traffic mode. The multiple-range analysis, however, showed that only the congested traffic emission pattern was significantly different from the other three, which led us to the discrimination of only two categories of traffic during a day. Table 3 gives the average carbon balance for the two categories we named congested and highway, the latter being the average of the three modes specified above. As already stated, the $P_i$ values from Tables 3 and 4 can be converted to emissions in g/km for tunnel traffic through multiplication by $\epsilon = 0.090$ kg C/km.

During the congested periods, an appreciable quantity of high speed traffic still is present because congestion generally occurred on two lanes at the end of the tunnel and gradually increased from there.
Table 3. Emission values $P_i$ in g/kg carbon in tunnel air for different traffic modes.

| Highway           | Congested |
|-------------------|-----------|
| Sulfur            | 3.38      |
| Carbon monoxide   | 4.4       |
| Carbon dioxide    | 3.39      |
| Nitric oxide      | 4.4       |
| Nitrogen dioxide  | 4.4       |
| Nitrogen          | 4.4       |
| Methane           | 4.4       |
| Ethane            | 4.4       |
| Propane           | 4.4       |
| Isobutane         | 4.4       |
| Butane            | 4.4       |
| Pentane           | 4.4       |
| Methyl-cyclopentane| 4.4     |
| Benzene           | 4.4       |
| 3-Methyl-hexane   | 4.4       |
| m- and p-Xylene   | 4.4       |
| o-Xylene          | 4.4       |
| Nonane            | 4.4       |
| Isopropylbenzene  | 4.4       |
| n-Propylbenzene   | 4.4       |
| 3-Ethyltoluene    | 4.4       |
| 1,3,5-Trimethylbenzene| 4.4 |
| 2-Ethyltoluene    | 4.4       |
| 1,2,4-Trimethylbenzene| 4.4 |
| Decane            | 4.4       |
| 1,2,3-Trimethylbenzene| 4.4 |
| Undecane          | 4.4       |
| Dodecane          | 4.4       |
| Tridecane         | 4.4       |
| Tetradecane       | 4.4       |
| Pentadecane       | 4.4       |

Table 4. Emission values $P_i$ for polycyclic aromatic hydrocarbons in g/kg carbon in tunnel air.

| Hydrocarbon        | $P_i$, g/kg C |
|--------------------|--------------|
| Pyrene             | 0.00028      |
| Chrysene           | 0.00018      |
| Benz[a]anthracene  | 0.00021      |
| Benz[a]fluoranthene| 0.00021      |
| Benz[b]fluoranthene| 0.00017      |
| Benz[o]fluoranthene| 0.00005      |
| Benz[e]pyrene      | 0.00018      |
| Benz[al]pyrene     | 0.00018      |
| Perylene           | 0.00003      |
| Indeno[1,2,3,c,d]pyrene| 0.00017 |
| Dibenzoanthracene[a,c]+[a,h] | 0.00002 |
| Benzog[b]pyrene    | 0.00035      |
| Anthanthrene       | 0.00003      |
| Coronene           | 0.00013      |

Sulfur to Carbon Ratio and Fraction of Diesel

The net carbon to sulfur ratio in tunnel air can be used to estimate the relative quantities of gasoline and diesel fuel used in the tunnel and the fraction of gasoline and diesel vehicles.

The calculation is based on the difference in sulfur content of these fuels in Belgium: gasoline has a maximum sulfur content of 0.05%, but on the average 0.01%, $S$ is present. Diesel fuel has a maximum allowable sulfur content of 0.2%, and this value is kept as close to this limit as possible by the refineries. Because the carbon content of both fuels is 0.863% and 0.861% kg/kg fuel, respectively, we can calculate their respective sulfur to carbon mass ratios as 0.12 and 2.3 g/kg.

In the polluted tunnel air, an intermediate $S/C$ ratio will be found. Figure 4 illustrates how the net sulfur to carbon ratio in the tunnel air is used to estimate the fraction of carbon from diesel by linear interpolation. Figure 5 shows the history of the net SO2 and CO2 concentrations for one day and the calculated fraction of carbon from diesel. The overall diesel/gasoline carbon ratio is obtained as the average weight by the hourly traffic density. It is obvious that the night observations, having a calculated diesel fraction of more than 100%, suffer from a systematic error that could not be properly identified yet. Either the tunnel sulfur concentration is raised during the nights by a sulfur source in the tunnel (e.g., SO2 desorption from the tunnel walls if the humidity at night rises) or the background SO2 levels are unexpectedly low due to some fast outdoor elimination mechanism. The reverse artifacts can be postulated for carbon dioxide, where a possible explanation would be a local CO2 source influencing the background measurement during the night. The total emissions in the tunnel can be subdivided into emissions from gasoline and diesel vehicles. This is accomplished by the equation:

$$P_i = P_i,g \times (1-F_d) + P_i,d \times F_d$$

where $P_i,g$ as $P_i$, from gasoline vehicles; $P_i,d$ as $P_i$, from diesel vehicles; $F_d$ is the fraction of carbon originating from diesel in emissions.

The values of $P_i,g$ and $P_i,d$ are calculated from Equation 4 by introducing their ratio from bench and road test emission factors (10) for the vehicle population and speeds prevailing in the tunnel. $P_i,d$ is calculated as the sum of emissions $P_i$ from trucks and diesel passenger cars, taking into account average emission factors and fuel consumption for both groups.

The splitting of emissions into categories is needed for the extrapolation of the tunnel data to national scale emissions, which have a slightly different pattern of vehicles and fuel use. In the tunnel, $P_i,d$ is the measured diesel carbon fraction, and the percentage of freight traffic was counted as 20%. For the calculation of the national emission budget, the statistics of fuel sales, or alternatively the vehicle population statistics with an estimated yearly mileage per vehicle class are used.

Estimates of Emissions from Tunnel Data

From the data for high-speed traffic in Table 3, highway emissions in g/km are...
Table 5. Estimated highway emissions in g/km.

| Component            | From tunnel data | CORINAIR model |
|----------------------|------------------|----------------|
| CO                   | 4.49             | 3.78           |
| NO                   | 4.01             | 3.99           |
| NMHC                 | 1.33             | 0.68           |
| Sum of volatile      | 0.40             |                |
| compounds analyzed   |                  |                |
| Benzene              | 0.023            |                |
| Toluene              | 0.055            |                |
| Ethylbenzene         | 0.011            |                |
| m+p-Xylene           | 0.032            |                |

Table 6. Emission estimates from tunnel emission factors compared to Flemish Region Emissions Inventory (EIVR).

| Component            | From tunnel data | EIVR |
|----------------------|------------------|------|
| CO                   | 184              | 519  |
| NO                   | 165              | 178  |
| NO₂                  | 0                |      |
| SO₂                  | 8                | 18   |

* Data calculated as kg/day x km of highway.

Table 7. Estimates of total yearly emissions in Belgium based on tunnel emission factors (ton/year).

| Highway       | Congested |
|---------------|-----------|
| CO            | 160000    | 285000 |
| NO            | 106000    | 95000  |
| NO₂           | 13100     | 12200  |
| NO₂ (as NO₂)  | 175000    | 159000 |
| SO₂           | 14500     | 14500  |
| Methane       | 0         | 0      |
| Ethane        | 6760      | 2150   |
| Propane       | 893       | 374    |
| Isobutane     | 301       | 306    |
| 1-Butene      | 423       | 251    |
| Butane        | 724       | 525    |
| Pentane       | 198       | 123    |
| Methyl-cyclopentane | 188  | 150    |
| Benzene       | 855       | 1260   |
| 3-Methyl-hexane| 213       | 334    |
| 2,2,4-Trimethyl-pentane | 284  | 512    |
| n-Heptane     | 213       | 150    |
| Toluene       | 2130      | 2950   |
| 3-Methyl-heptane | 55       | 124    |
| Octane        | 91        | 106    |
| Ethylbenzene  | 428       | 586    |
| m and p-Xylene| 1170      | 1890   |
| o-Xylene      | 564       | 624    |
| Nonane        | 117       | 203    |
| Isopropylbenzene| 13        | 26     |
| n-Propylenzene| 142       | 199    |
| 3-Ethyltoluene | 851       | 1124   |
| 1,3,5-Trimethylbenzene | 165  | 218    |
| 2-Ethyltoluene | 187       | 245    |
| 1,2,4-Trimethylbenzene | 507  | 716    |
| Decane        | 219       | 488    |
| 1,2,3-Trimethylbenzene | 164  | 250    |
| Undecane      | 175       | 428    |
| Dodecanec     | 126       | 226    |
| Tridecane     | 104       | 191    |
| Tetradecane   | 107       | 141    |
| Pentadecane   | 98        | 99     |

Table 8. Estimates of traffic polycyclic aromatic hydrocarbons emissions from tunnel emission factors.

| Components  | Ton/year | µg/km |
|-------------|----------|-------|
| Pyrene      | 1.06     | 25    |
| Chrysenene  | 0.69     | 18    |
| Benzo[a]anthracene | 0.82 | 19    |
| Benzo[k]fluoranthene | 0.81 | 19    |
| Benzo[a]pyrene | 0.64     | 15    |
| Benzo[a]pyrene | 0.19     | 5     |
| Benzo[b]pyrene | 0.68     | 16    |
| Benzo[a]pyrene | 0.69     | 15    |
| Benzo[b]pyrene | 0.11     | 3     |
| Indeno[1,2,3,c,d]pyrene | 0.65 | 2     |
| Dibenzo[a,h]anthracene | 0.07 | 15    |
| Benzo[p,h]perylene | 1.33     | 32    |
| Anthanthrene | 0.11     | 3     |
| Coronene     | 0.51     | 2     |

calculated for a traffic speed of 120 km/hr in Table 5, and compared to the results obtained with the emission factors given by CORINAIR (10) at the same speed.

It is apparent that the agreement between the tunnel data and the CORINAIR emission estimates is good for CO and nitrogen oxides, while for nonmethane hydrocarbons, almost twice the emission predicted by CORINAIR is obtained. The underestimation of CO and hydrocarbon emissions by models is a well-known fact and this is generally blamed to a limited number of badly tuned engines with extremely high emissions. The degree of underestimation by CORINAIR therefore is not excessive compared to some (27). The emission factors used in 1990 by Flanders Region Emissions Inventory, the official body that makes the emissions inventory in the region of Flanders, where the tunnel is situated, give quite deviating results from ours for CO and SO₂, while for NO, there is good agreement again (Table 6). The estimates in Table 6 were obtained with specific emission factors (Equation 4) and vehicle counts in the tunnel. The results reported are averages of 3 days.

Table 7 lists 1991 national emission estimates for Belgium, based on the application of the carbon balance to the tunnel data, the fuel sales statistics, and the motor vehicle population. The tunnel measurements represent only two traffic modes, and items like cold starts are not included. High speed driving is certainly overrepresented and therefore we assume that the Congested column in Table 7 is the best approximation for the nationwide emissions. The PAHs emission estimates given in Table 8 are based on 24-hr averages and thus represent the average traffic situation like it exists in the tunnel.

An already cited advantage of emission estimates by the carbon balance method is that a real sample of vehicles is taken on the road in realistic driving conditions. Our measurements involved a sample of 23,000 to 47,000 vehicles per day. A further advantage is that emission factors for components that are otherwise unavailable can be obtained readily.

The uncertainty in the nighttime determination of the diesel fraction from the sulphur and carbon content demonstrates some inherent weaknesses in the carbon balance method. Therefore, we can formulate the following conditions for its applicability: a) The accurate determination of background concentrations is of utmost importance. Whenever possible, simultaneous measurements are advisable. b) The pollutant concentrations in the tunnel must be elevated to eliminate precision errors when background is subtracted. A tunnel with efficient ventilation and low traffic density is a less desirable object for this purpose. c) It must be assumed that no significant deposition or absorption of components takes place in the tunnel. During wet weather conditions sulfur dioxide is found to disappear from the tunnel by wash out (11). Aerosols, especially soot, are visibly deposited on tunnel walls. d) It must be possible to take representative samples of all pollutants of interest. This implies that the tunnel air must be well mixed and that eventual particle phase pollutants behave like gases, and if they behave otherwise, they are adequately taken care of during sampling. Which particle size cut to use (e.g., total suspended particles or PM10, particulate matter below 10 µm) may have another answer in a tunnel environment than in ambient air. Further experimental study is required to optimize aerosol sampling in traffic tunnels.

The Lead Balance as a Test Case

The gradual reduction of lead concentrations in gasoline from 2 g/L and higher to 0.15 g/L has caused a spectacular decrease of ambient air lead levels in Belgian cities. The use of unleaded gasoline and exhaust catalysts in significant portions in Belgium began in 1989. In January 1991, unleaded accounted for 25% of gasoline sales, and in March 1992 this figure had risen to 45%. The preliminary 1991 figures of transport fuel sales in Belgium, obtained from the Ministry of Economic Affairs, are 3,546,564 tons of diesel and 2,700,333 tons of gasoline. Gasoline sales are subdivided as follows: 15,838 tons of normal unleaded (92 octane), 794,921 tons of Eurosuper (95 octane), 206,639 tons of Super (98 octane), 1,682,935 tons of Super Leaded (0.15 g/L Pb).

Emission regulations state that new gaso-
line cars with displacement above 2000 cc need to be equipped with three-way catalysts from January 1990 onwards. From July 1990 until July 1992 smaller cars benefit from a bonus system that partially refunds catalysts on new cars. In practice, this has resulted in low penetration of catalysts in this category. By July 1992, all new types, and by January 1993, all new cars with gasoline engines will have to be equipped with three-way catalysts in order to comply with the European Economic Community directive on emissions 91/441/EEC.

These fast changes in regulations undoubtedly will result in an important decrease in traffic emissions, including the complete elimination of lead. The emission of lead by gasoline cars in the tunnel offers an interesting test of the carbon balance principle on an aerosol-bound pollutant.

The theoretical quantity of lead released in the tunnel can be estimated with reasonable accuracy, and the result can be compared to the actual measurements. Table 9 gives measured lead concentrations in the tunnel and the corresponding 24-hr average increase of carbon dioxide concentrations.

The background lead concentration in the air in this area is monitored by the IHE with the same equipment we used in the tunnel. As a yearly average for 1989 to 1990 in the nearby heavy metal monitoring station HOB04, the value of 0.19 µg/m³ was measured. Therefore we calculate a lead to carbon ratio (Pb/C) in the tunnel air of 0.0143 g/kg. However, the expected Pb/C ratio, based on a 44% fraction of carbon from gasoline, a 62% fraction of leaded gasolines, and a lead content of 0.15 g/L is 0.0652 g/kg. This means that only 22% of the expected lead is recovered. This surprising result prompted us to treat the data obtained in two Brussels tunnels in 1989 by the IHE (11) in a similar way. These measurements represent a situation with relatively more passenger cars and a lower carbon fraction from diesel $F_d = 17.4%$. The reported net lead concentrations were 5.832 and 4.65 µg/m³, respectively. Taking into account the carbon content of these tunnel atmospheres, Pb/C ratios of 0.0478 and 0.04 g/kg are found. With 84.6% leaded gasoline used at that time, the lead recovered by the samplers is 28.6 and 24% of the lead in the gasoline, respectively. It is known that lead emissions from vehicles are entirely in the particulate phase and that this freshly emitted aerosol is very fine, with aerodynamic diameters below 10 µm. Therefore, we suspect no sampling artifacts and must assume that actually only 22 to 29% of the lead in gasoline is emitted. This contradicts the assumed CORINAIR lead emission factor of 75%. However, it is our opinion that this issue needs further clarification before the carbon balance can be applied to aerosol related pollutants with confidence.

**Conclusions**

The mass balance over a traffic tunnel by means of the carbon balance offers a relatively fast way of assessing on-the-road emissions for a large number of cars under actual driving and maintenance conditions. The method is useful to check the available emission factors and emissions inventory systems. Furthermore, it can be used to determine traffic-caused emissions of pollutants for which no data are available presently. However, care must be taken to avoid artifacts during tunnel and intake air analysis and some assumptions must be made in applying the method. Its applicability to aerosols still requires further validation.

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