Definition of PM₁₀ emission factors from traffic: use of tracers and definition of background concentration

E. Brizio & G. Genon *Turin Polytechnic, Italy*

Abstract

Within air quality management, one of the most important emission factors that should be known is PM_{10} (exhaust and non-exhaust flows) coming from traffic. The existing data and models produce very variable emission factors, also according to climate, sanding conditions, road material; thereby, a more general approach, based on the use of traffic tracer such as CO and NO_x , can be put into practice in order to have a reliable assessment of PM emissions. Within the tracer approach, the definition of the background concentration of pollutants is of prime importance but representative measurements could be not at disposal. The present work is an attempt to define background concentration by considering the average concentration measured in an urban area during the night (0-5 am), when traffic and industrial instantaneous contribution are negligible and the heating plants are switched off. The so called "night method" has been tested and validated by means of the OSPM model, an atmospheric dispersion model studied for street canyons, for CO and NO_x. In the case of CO, the results were surprisingly satisfactory and the method could be considered consistent, whereas NO_x turned out to be not reliable as a tracer because of the chemical reactions that occur in the troposphere.

Keywords: traffic, air quality, atmospheric modelling, background concentration, OSPM, CO, PM_{10} , NO_x , tropospheric chemistry.

1 Introduction

The air pollution situation of many European urban areas doesn't present indications of substantial improvement, in spite of the adoption of specific policies of limitation and emission reduction; actually, these actions, without



other activities, like clear understanding of emissive and atmospheric phenomena influencing the result, are not able to lead the air quality back to desired standards. The air quality situation is even more critical in areas like Northern Italy, where the pollution levels (in particular PM_{10} and NO_2) are very high because of the low wind conditions of the Po Valley that don't help the dilution of the pollutants. In order to obtain some improvements for air quality, the regional decision makers are trying to define some intervention policies, such as the limitation of old vehicles, in particular diesel cars before EURO II and gasoline cars before EURO I. The present paper deals with PM emissions from traffic, considering exhaust and non-exhaust particles. The investigated area is the town of Cuneo, 50,000 inhabitants, placed in the South of Piedmont, N-W Italy. The mean wind speed in the area is quite low, around 1.4 m/s, with an high percentage of calm hours (< 1 m/s), almost 30%, and a typical bimodal behaviour around 40-60 degrees clockwise from the N.

Traffic flows for all the main street of the town, registered by magnetic counters, account for more than 308,000 vehicles per day.

2 PM emissions from traffic

PM emissions from traffic can be divided into three main groups (Ketzel et al. [1]):

- direct exhaust emissions, mainly fine fraction (PM_{2.5}), that can be calculated by means of different emission databases (i.e. COPERT, UBA, TNO, CORINAIR, UK-TLR);
- non-exhaust emissions deriving from brakes wear (PM₁₀-PM_{2.5});
- non-exhaust emissions from road abrasion, tyre wear and road dust resuspension that are found partly in the fine fraction $(PM_{2.5})$ and mostly in the coarse fraction (PM_{10}) .

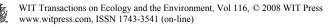
PM emissions are strongly influenced by external factors as road condition (wetness, salting, sanding, road material) and use of studded tyres.

Literature data reports several different model to define in particular nonexhaust emissions that can be very variable, as pointed out by Figure 1.

As one can easily understand, the provided database are quite variable and, most of all, they have been obtained in correspondence to precise conditions of weather and road characteristics that are strongly site specific. A more general and reliable approach could be the so called "tracer method", used within the Swedish Empirical Model [2] in order to obtain the total PM emission factor, including both direct emissions and emissions from the dust layer. The method can be written as follows, using for example CO (or NO_x) as tracer:

$$e_{f}^{PM} = e_{f}^{CO} \cdot \left(\frac{C_{PM}^{roadside} - C_{PM}^{background}}{C_{CO}^{roadside} - C_{CO}^{background}} \right)$$

where ef^{CO} is the emission factor for CO (or NO_x), often more well known than the PM one.



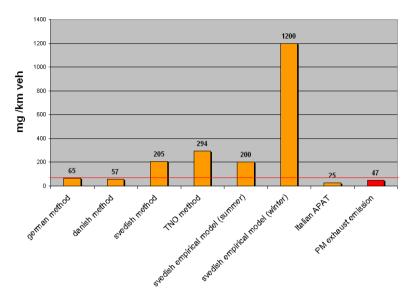


Figure 1: Non-exhaust PM₁₀ emission factors (fleet mix).

3 Background concentration and stagnation phenomena

As already described, the "tracer method" needs the definition of the background concentration of the involved pollutants, a very critical parameter. The background pollution level is usually measured in the countryside or at a rooftop within a urban context. In the analyzed area, all the monitoring stations are placed in urban areas and the measured values are almost the same. So we don't have any background monitoring station at disposal for our purposes. Moreover, on the basis of our experience, the background concentration that can be measured in the countryside is not the same as the one that can be measured in a urban environment; this can be observed, for example, when the traffic is totally stopped for sanitary reasons (the so called "no traffic Sundays") in the cities. This aspect is quite reasonable if one considers that the background concentration is also due to a stagnation effect of the pollutant emitted in the previous hours only partially dispersed by the wind and the atmospheric turbulence (mechanically and thermally induced); this way the background concentration is strongly dependent on the emission mixture and the dispersion capabilities of the area. For instance, the background concentration measurable in a street canyon would be mostly correlated to traffic as the main emission source and it would be probably higher than that measured in an outside area (or also in a urban background station placed on a rooftop, as indicated by Oemstedt et al. [2]) because of the low dispersion possibilities of a urban canyon if compared to a more open area. The described assumption can be better explained by Figure 2, taken from Hansson [3].



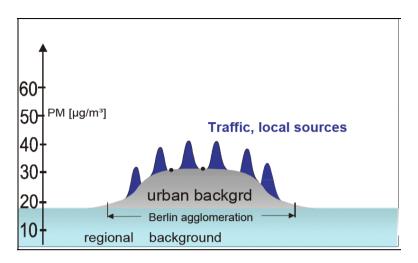


Figure 2: Example of urban background concentration.

The difference between the background concentrations measured in a rural site, at a rooftop or at the bottom of a street canyon has been also observed in [4]. There, one can also notice that during the night the three measured levels are very close, so that this level seems to be a good approximation of the representative background concentration of the studied area.

Based on the reported arguments, we tried to define the background concentration for stable parameters, such as CO and also PM_{10} , by calculating the average concentration from 0:00 am to 5:00 am, when the traffic is low in small towns and the heating plants are not working; moreover, the instantaneous concentrations at the ground level due to industrial plants are very low, so that we assumed those contributions as negligible in this phase. The described way to define the background concentrations, called "night method", can be considered valid for pollutants such as CO or PM_{10} , that are quite stable in atmosphere (the lifetime is respectively in the order of months and weeks), but an attempt will be carried out also for NO_x .

Once the background concentration has been defined, the procedure can be validated by means of reliable atmospheric dispersion models that can predict the instantaneous effect of traffic (and the other sources) on air quality. This contribution can be added to the background concentration and compared to the measured daily concentration. In order to simulate the effects of traffic emissions on the urban air quality we used the Operational Street Pollution Model (N.E.R.I., Denmark [5]).

Figure 3 reports the comparison of measured and modelled CO daily concentrations at the monitoring station of Cuneo. As one can easily observe, the modelled values reproduce the measured one in a satisfactory way, the correlation coefficient is very high (r=0.977), so that the model and the approach can be considered reliable for our purposes.

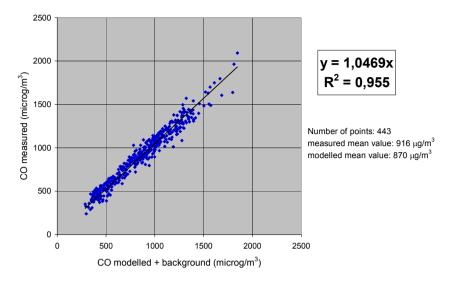


Figure 3: Comparison of measured and modelled CO daily mean concentrations in Cuneo.

It should be remembered that, as far as the CO emissions deriving from the heating plants and the industrial activities are concerned, their effect on air quality can be considered around 5 μ g/m³, as maximum daily concentration at the ground level; the reported levels is negligible if compared to the CO concentrations measured in the analyzed area (300-2000 μ g/m³).

As a consequence, we may say that the instantaneous effect of sources, other than traffic, is very low on the air quality of the analyzed area and so it is acceptable to neglect them when applying the described method, as we assumed in the present paper. This reasoning can be extended to other pollutants as well, such as PM_{10} and NO_x , in the studied region.

4 The background concentration for unstable parameters: the case of NO_x in Southern Piedmont

As already pointed out in the previous chapters, the "night method" turned out to be a reliable tool in the case of stable parameters such as CO and PM_{10} . Nitrogen oxides (NO + NO₂) are considered and used as tracer of traffic emissions as the sum of NO (as NO₂) and NO₂ remain constant within the photolytic cycle.

The time scales characterizing these reactions are of the order of tens of seconds; in the case of residence time of pollutants in a street canyon comparable to the mentioned reaction time, the chemistry of NO_x could be restricted to the described cycle [6]. As a consequence, the "night method" applied to the sum of NO_2 and NO (calculated as NO_2) should be theoretically viable in order to obtain the background concentration.

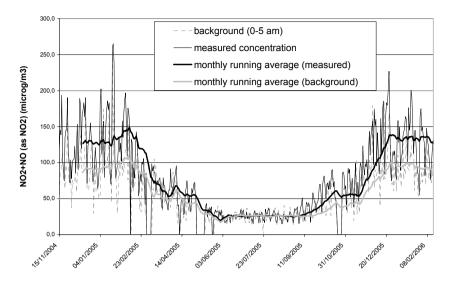


Figure 4: Measured and background daily concentrations for NO_x in Alba.

Thereby, we applied the same approach described in chapter 3 for carbon monoxide to NO_x for several urban contexts in Southern Piedmont (Cuneo, Alba, Bra, Asti). Here we found out an unexpected behavior of the NO_x background concentration calculated by means of the "night method", as pointed out by Figure 4. As a matter of fact, during the summer the night average concentration of NO_x is equivalent to the daily average concentration, and this can be observed for all the analyzed monitoring stations. The unreliability of the "night method" applied to unstable parameters is confirmed by the prediction of overall daily concentration (background + traffic instantaneous concentration calculated by OSPM at the monitoring station placed in Cuneo), reported in Figure 5. In this case, we won't find a satisfactory correlation, on the contrary a clear overestimation of the measured concentrations can be observed.

The behaviour of NO_x background concentration during the warm season seems to suggest that the sum of NO (as NO_2) and NO_2 is not constant within the residence time of these pollutants in the street canyon, on the contrary, other complex reactions involving a removal or a production of NO_x may occur, respectively during the daytime or at night. In this case, the presence of other chemical mechanism (in addition to the photolytic cycle of NO_x) with reaction times longer than seconds could be justified by large residence times of pollutants inside the street canyon, due to the stagnation phenomena within the Po basin.

The phenomenon has been analysed by comparing the daily trends of NO, NO_2 and O_3 during the year and the summer at some monitoring stations placed in the South of Piedmont (Brizio and Genon [7]). The study seems to suggest that, during the day, NO is consumed without being transformed in NO_2 and also NO_2 appears to be removed by chemical reactions different from photolytic

cycle (that should maintain constant the sum of NO_x). Anyway, during the summer the trends of NO_x don't show the influence of daytime emissions, unlike the yearly behaviour, confirming the presence of a "sink".

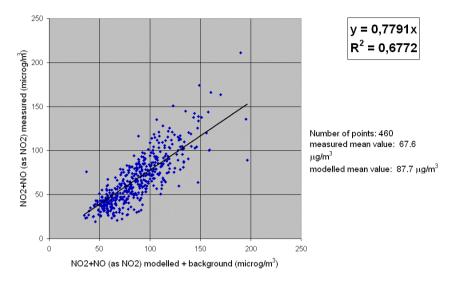


Figure 5: Comparison of measured and modeled NO_x daily mean concentrations in Cuneo.

5 Definition of PM₁₀ emission factors by means of tracers

As described in chapter 2, pollutants emitted by traffic such as CO and NO_x can be used as tracer to define emission factors with an higher level of uncertainty, signally PM₁₀. Figure 6 points out PM₁₀ emission factors (exhaust + nonexhaust) obtained in Cuneo by using CO and NO_x as tracer; as one can notice, the data are quite similar during the cold seasons, while during the summer the emission factors obtained through NOx tends to be strongly overestimated (see also Figure 7). As already described, NO_x is not as stable as the other pollutants, CO and PM₁₀, and this behaviour lead to a wrong definition of the reference NO_x concentrations to be run within the "tracer model". As a consequence, its use as a tracer is at least questionable.

On the contrary, CO turned out to be suitable for a reliable application of the "night method" for the background definition and the atmospheric dispersion modelling so that its use as a tracer seems to be the most adequate choice (Brizio et al. [8]). The calculated PM_{10} emission factor, as obvious, changes according to the season and the wetness of the atmospheric conditions. The emission factor calculated using CO as a tracer varies around a mean value of 257 mg/km/veh \pm 164 mg/km/veh, with a maximum value of 1136 mg/km/veh; the reported data are much higher than the values referred by the "German method" and the "Danish method" while we can find a good agreement with the CEPMEIP-TNO



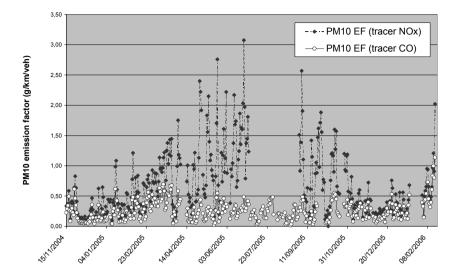


Figure 6: PM₁₀ emission factors calculated by using CO and NO_x as tracers.

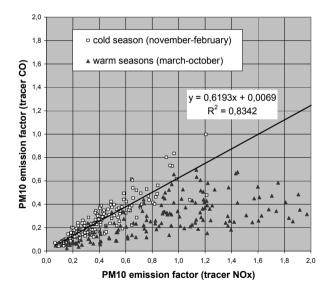


Figure 7: Comparison of calculated PM₁₀ emission factors.

suggested data and most of all with the Swedish values, in particular the described range 200-1200 mg/km/veh.

Based on the PM_{10} emission factors obtained by means of the "tracer method" and the OSPM model, we calculated the PM_{10} concentrations, as reported in Figure 8. Also in this case the correlation is very good (r=0.959), even though

the model lightly underestimate the measured concentration. The mean deviation D, defined as follows:

$$D = \sum_{i=1}^{n} \frac{\left|Cm_{i} - Cc_{i}\right|}{n} \cdot \frac{1}{\overline{Cm}} \cdot 100$$

where Cm is the measured concentration and Cc is the calculated concentration, is less than 17%.

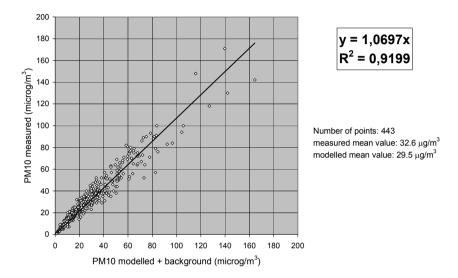


Figure 8: Comparison of measured and modelled PM₁₀ daily mean concentrations.

6 Conclusions

 PM_{10} emissions from traffic is a very important parameter when dealing with emission inventories, air quality management and policy making. Unfortunately, its definition is not straightforward as it depends on several site specific parameters so that an assessment based on tracers within the area of interest seems to be advisable. In this context, not all the pollutants emitted by vehicles are suitable for a reliable use of the "tracer method". In particular, NO_x is a very complex parameter and the prediction of its behaviour may be very complicated. Based on this conclusion, the application of the "night method" in order to define the NO_x background concentration could be not advisable, at least during the summer. More generally, in the analyzed area, the use of NO_x as a tracer for traffic could be questionable as well, because of the rapid chemical reactions that can occur also at time scales characterizing the dispersion of pollutants within a street canyon. On the contrary, the use of CO, a stable and easily predictable parameter, could lead to consistent results: in the studied area we obtained total



PM emission factors varying around a mean value of 257 mg/km/veh \pm 164 mg/km/veh, with a maximum value of 1136 mg/km/veh, while the mean exhaust PM emission factor calculated by means of the Copert3 model is 47 mg/km/veh. The described methodology indicates that 80% of the total emitted PM₁₀ originates from non-exhaust emissions and so it is evident that policies reducing the exhaust releases of the park or limiting diesel vehicles without particle traps can have a limited effect on the air quality.

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