FROM EMISSION TO IMMISSION: THE WAY TO PERTINENT EVALUATION OF TRANSPORT-RELATED HEALTH AND ENVIRONMENTAL IMPACTS

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ABSTRACT
This contribution focuses on emission/immission contribution assessment of road transport to global pollution source apportionment. The recent evolution in this domain show the impressive reduction of tailpipe emissions when brake and tyre wear PM emissions become the major source of recent powertrains. Inside vehicle immissions have been assessed using a vehicle inserted in the traffic as a ‘mobile laboratory’. These immissions may represent the major part of the daily personal space-time exposure budget to air pollution. As an example, each hour spent in a car cabin may represent to a 1 hour exposure above the WHO recommendation limits for both PM and NO2. Time course evolution of car cabin immissions shows an important reduction of PM exposure thanks to the Euro5 regulation which made mandatory the use of particulate filters on light duty diesel-powered vehicles. In the meantime, recent toxicological studies demonstrate the impressive reduction of genotoxicity, carcinogenicity, and pro-inflammatory potentials of post Euro5 diesel-vehicle emissions. As a conclusion, the impressive reduction of post Euro5 diesel-vehicle tailpipe emissions leads to a reduction of traffic-related pollutant immissions which will further improve with the expected growing of Euro6 complying vehicles in the forthcoming years.

Keywords: atmospheric pollution, emissions, health effects, immissions, nitrogen oxides, particulate matter, personal exposure, space-time exposure budget traffic.

1 INTRODUCTION
Road transport tailpipe emissions and more specifically diesel engine-powered vehicle emissions are most frequently claimed as being the major source of city air quality degradation. Diesel engine-powered vehicles. In this respect, WHO classified diesel engine-emitted particulate as class 1 carcinogen to humans in 2012 [1], and atmospheric pollution as class 1 carcinogen to humans in 2013 [2]. Tailpipe particulate matter emissions especially from diesel engines have been tremendously reduced due to the onset of Euro5 regulation on engine emissions that rendered mandatory the use of closed particulate filters on the exhaust lines for diesel engine-powered vehicles produced since year 2011. This very efficient (over 95%) reduction of diesel soot emissions led to take into account the friction particulate emissions from brake wear and tire wear as becoming a new concern from vehicle powertrain emissions. We will consider three sections in this contribution: the first section will be dedicated to the assessment of particulate matter source apportionment during seasonal particulate matter pollution episodes, the second section will be dedicated to the assessment of daily personal exposure budget with a specific approach of car cabin exposure compared to other transportation choices. Finally, the third section will be dedicated to recent time course evolution of car
cabin immission and the extensive reduction of diesel-engine emission genotoxicity and inflammatory potentials according to emission after-treatment strategies.

2 ATMOSPHERIC POLLUTANT SOURCE APPORTIONMENT

An overview of the French pollutant emission inventory published by CITEPA [3] shows a constant reduction of pollutant emissions since at least year 1990. Figures 1–3 summarize this observation for particulate matter, NOx, and non-methane VOC.

It is of interest that for these pollutants, reduction of road transport contribution by 90% for NMVOC, 60% for NOx, 70% for PM1, is one of the most important triggers responsible for the improvement of global emission reduction thanks to the advanced technologies in the field of engine exhaust after-treatment namely there way catalysis on spark-ignited engines and oxidation catalysis and particulate filters on diesel engines. As shown in Figs 4 and 5, these improvements remain highly significant when expressed per inhabitant and or per ton equivalent petroleum thus confirming the improvement achieved in the field of anthropic source depollution strategies.

Since year 2007, particulate matter monitoring by air quality networks is performed using the TEOM/FDMS technology including the semi-volatile fraction of particulate matter which was not formerly included when using the TEOM technology. This change in particulate assessment technology had a very significant impact on particulate matter levels especially during spring particulate matter pollution episodes where a high semi-volatile fraction made essentially of ammonium nitrate and ammonium sulfate is observed as shown in Fig. 6 and described in Rouil et al. [4].

In year 2011, particulate matter threshold have been significantly reduced from 120 to 80 and from 80 to 50 µg/m³ for population alert and information, respectively. Both actions have been responsible for an artificial increase of the number of alerts while in the meantime

![Figure 1: PM1 emission source apportionment for France. kton/year. Data calculated from [3]. NTR non road transport, TR road transport, AS Agriculture and forest, RT residential and tertiary, IM Industry Manufacture, TE Energy transformation.](image-url)
atmospheric particulate matter concentrations significantly decreased as reported by AirParif [5] and the French minister of environment [6] although some yearly very typical PM episodes are observed in March. Interestingly enough, these particulate matter pollution episodes observed during early spring periods since the introduction of TEOM/FDMS metrology would not have been considered as such when using TEOM metrology only since solid particles did not exceed the 50 µg/m$^3$ threshold (Fig. 6). During these episodes, a very peculiar

Figure 2: NOx emission source apportionnement for France. kton/year. Data calculated from [3]. NTR non road transport, TR road transport, AS Agriculture and forest, RT residential and tertiary, IM Industry Manufacture, TE Energy transformation.

Figure 3: Non methane VOC emission source apportionnement for France. kton/year. Data calculated from [3]. NTR non road transport, TR road transport, AS Agriculture and forest, RT residential and tertiary, IM Industry Manufacture, TE Energy transformation.
elevation of PM10 volatile/solid PM ratio which is not occurring during other periods of the year. These episodes are not at all followed by any significant health impacts such as increased hospital admissions, asthma or bronchitis exacerbation Host et al. [7]. Particulate matter chemical composition as assessed by the French national program CARA appears to be very
specific during these episodes, with a very high contribution of nitrates, ammonium and sulfate which may exceed 50% of PM10 and account for the vast majority of volatile PM10 fraction [8].

Concerning epidemiology studies and elaboration of European recommendations for population information and alerts, it is all the most important to take into account that most of the European relative risk data still in use today were elaborated in the frame of the APHEA [9, 10], APHEIS [11] and APHEKOM [12] projects which took place before the implementation of air quality monitoring networks with the TEOM-FDMS. Data for PM measurements were thus not including the volatile fraction of PM. In view of these considerations, one may question the potential interest of the inclusion of the volatile fraction of PM for the onset of PM information and alert procedures at least during the spring PM10 episodes which represent ca. 90% of the yearly observed PM10 episodes in the north of France.

3 DAILY PERSONAL SPACE-TIME EXPOSURE BUDGET EVALUATIONS

Usual air quality/health impact epidemiologic studies are related to outdoor urban air pollutant concentrations provided by air quality monitoring networks. However, it is well-known that major significant variations of personal exposure levels occur with different environments to be visited, as well as the significant increase of personal exposure levels associated with some activities along a day. This has led epidemiology scientists to elaborate space-time-budget studies. In a first approach, they have considered the time spent at home as an important data to estimate exposure to indoor pollutants. The average daily time spent in the home was 16 hours and 10 minutes. This represents 67% of the daily time (71% for women and 63% for men). Data can be collected from French observatory of indoor air quality 2005.

Figure 6: Typical example of volatile and solid PM10 fractions during early spring PM10 episodes in the north of France. PM episode of March 2014 in Rouen city, calculation made from AirNormand monitoring network data. Solid: measurement with TEOM, Volatile: TEOM-FDMS minus TEOM measurement, total stack: TEOM-FDMS measurement. Values are 24 hour mean values.
In this study, we show that cabin air quality is a specific environment which may contribute very significantly to the daily space-time-budget of exposure to pollutant. In large cities, for daily commuting or for worker categories as taxi or lorry drivers, time spent in the traffic may

Figure 7: In car cabin, NO₂ concentrations according to the driving situation. Exposilla study [14]. Data are presented as box plots (centiles 5, 25, 50, 75, and 95) of recorded data during 4500 km for Global. Measurement were every each second with two analysers (AC32M and T32M Environnement SA France)) to cover the high dynamic range of concentrations encountered.

Figure 8: In car cabin PM1 concentrations according to the driving situation. Exposilla study [14]. Data are presented as box plots (centiles 5, 25, 50, 75 and 95) of recorded data during 4500 km for Global. Measurements were every each second using the conversion of ELPI (Dekati Finland) total current values.
last from 1 to 10 hours. Since about 10 years from now, we started to study the evolution of pollutant concentrations namely NO$_2$, NOx and particulate matter in the cabin of cars inserted in the traffic [13, 14]. We clearly demonstrated that extremely high PM and NO$_2$ concentrations were encountered in this confined area due to the situation of the vehicle in the exhaust plume of the preceding vehicle. As shown in Figs 7 and 8, few differences were observed between traffic situations such as urban, peri-urban highways. As a short summary of measurements every each second during more than 4500 km in 2007–2008, each hour spent in a vehicle inserted in the traffic may represent an exceed of WHO recommendation limits for exposure to both NO$_2$ and particulate matter. These concentrations are 3 to 5 times as big on average as those measured by air quality network on road side.

4 TIME COURSE EVOLUTION OF CAR CABIN IMMISSIONS

These observations clearly show that car cabin is one of the most polluted environments for general population exposure. This environment should thus definitely be taken into account for personal space-time budget evaluations since we could estimate that 1 hour spent in a vehicle might represent as much as 50% of total personal daily exposure to either NO$_2$ or particulate matter. Major time course evolution of car cabin NO$_2$, NOx, and PM concentrations has been observed over the last decade: Figs 9–11 show that using similar measurement protocols, in car cabin NO$_2$, NOx and PM concentrations significantly increased from year 2007 to year 2010. This increase was followed by a significant reduction from year 2010 to year 2015, with a more extensive decrease for PM1 than for NO$_2$ or NOx. These variations can be explained by the introduction of particulate filters in year 2011 with the Euro5 regulation on diesel personal cars and light duty vehicles. This introduction of very efficient particulate filters allowed the use of increased exhaust gas recirculation as a strategy for NOx reduction to comply with the Euro5 regulation which was also more stringent than Euro4 regulation for NOx emission.

Figure 12 shows that The PM/NOx ratio continuously decreased from 2007 to 2015 as a result of technological depollution strategies rendered necessary to comply with the Euro

![Figure 9: Time course evolution of car cabin NO$_2$ concentrations over the last decade. T inside tunnel concentrations, NT outdoor concentrations. Data are presented as box plots (centiles 5, 25, 50, 75, and 95).](image)
regulation evolution. It is expected to see a further reduction of both NO\textsubscript{2} and NO\textsubscript{x} emissions in the forthcoming years as a result of Euro 6 regulation complying vehicles coming to the market since October 2015.

5 CONCLUSIONS
We could demonstrate that the specific environment inside the vehicles inserted in the traffic is a highly polluted area which may contribute to 50\% of the daily space-time exposure.
budget. This environment pollutant immission is directly related to the quality of the vehicle exhausts in the close vicinity more than to the general outdoor pollution levels. Interestingly enough, the improvement of engine emission after-treatment strategies have a direct impact on the improvement of car cabin air quality and of the air quality in the proximity of the traffic as shown by our measurement and measurements from air quality monitoring networks, respectively, during the last decade. These observations on real life immission due to traffic emissions are very important to calibrate inhalation toxicological studies with pertinent aerosol dosimetry to the biological targets as conducted in our laboratory with either in vitro or in vivo models exposed for several hours to continuous flows of aerosols with controlled physicochemistry to assess acute biological responses. [15–17]. These observations may also be very important to help decision makers for the choice of the most suitable solutions to fight against specific pollutant emission sources owing their actual contribution to pollution apportionment and specific health impacts facing the variation of seasonal pollution episodes.

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