

# IMPLEMENTATION AND APPLICATION OF A SOURCE APPORTIONMENT APPROACH IN THE SIRANE URBAN AIR QUALITY MODEL

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**Abstract:** Ambient air pollution is the cause of significant effects on environment and health, leading to cancer and premature death of 7 millions people per year (WHO, 2014). This risk is especially high in the large European cities, where the concentration levels are important and often higher than those recommended by European regulations. To improve the life quality of population, it is necessary to reduce the concentrations of pollutants to acceptable levels. In order to develop effective strategies for reducing the concentrations, it is important to be able to determine the main origins/sources of the pollution. It is quite impossible, except for particular chemical species, to evaluate by measurements the contributors to the pollution at a specific location and time. On the other hand, the development of source apportionment modules in atmospheric transport and chemistry models allows to quantify the contribution of a specific emission sector, of a specific area or of a specific period of time. In this study, the contribution of the sources is determined by using the SIRANE urban air quality model (Soulhac et al., 2012, 2011). The method used for the source apportionment is based on the 'tagged species' approach, which consists on following the species emitted by the different groups of sources considered in the simulations. The difficulty in this type of approach is the treatment of non-linear phenomena such as chemical reactions. In this work, two different approaches have been tested and compared for the source apportionment with chemical reactions. The methodology, implemented within the SIRANE model, is then applied to estimate the average annual traffic, industrial, tertiary and background contributions on the concentrations of NO<sub>2</sub> in the agglomeration of Lyon for the 2008 year.

**Key words:** *Dispersion modelling, urban scale, source apportionment*

## **Introduction**

Ambient air pollution is the cause of significant effects on environment and human health, leading to cancer and premature death of 7 millions people per year (WHO, 2014). This risk is especially high in the large European cities where the concentrations levels are important and often higher than those recommended by the European regulations. In order to improve the life quality of the population, it is necessary to reduce the concentrations of pollutants to acceptable levels. In order to develop effective strategies for reducing the concentrations, it is important to be able to estimate the main origins/sources of the pollution. The source contribution can be estimated by means of monitoring measurements (for the particulate matter) and/or by means of numerical atmospheric dispersion model. The objective of this study is to compare two source apportionment models at the urban scale with the SIRANE urban air quality model (Soulhac et al., 2012, 2011).

## **Description of the SIRANE model**

SIRANE is an operational model which represents the pollutants dispersion at the urban scale. This model, based on the street network concept proposed by Soulhac (2000), uses a simplified representation of the urban geometry and adopts parametric equations to model the main flow phenomena in the urban area: advection along the street axis induced by the parallel component of the wind; turbulent diffusion across the interface between the street and the external atmosphere; exchanges at the street intersections. The roughness sub-layer just above the urban canopy is neglected and the flow in the external atmosphere is modelled as a boundary layer flow on a rough surface. This external flow is supposed to be horizontally

uniform. In the external atmosphere, the pollutants dispersion is represented by means of a Gaussian plume model with the standard deviations parametrized by the Monin-Obukhov similarity theory. The SIRANE model estimates the hourly mean concentrations assuming stationary conditions at each time step. The input data are the urban geometry of the streets network, the meteorological data, the point, line, and distributed emissions and the background concentration coming from the exterior of the domain.

The chemical reactions taken into account in the SIRANE model are relative to the NO-NO<sub>2</sub>-O<sub>3</sub> cycle. These reactions occur at a time scale very much inferior to the hourly time step used by the SIRANE model (Seinfeld, 1986). Consequently, it is important to model these reactions in order to take into account the concentrations modifications they induce. Those are modelled with the Chapman cycle (1) where  $k_1$ ,  $k_2$ , and  $k_3$  are the kinetic constants of reaction.



The radical  $\text{O}^*$  being very reactive, the characteristic time of the second reaction leading to production of ozone is negligible in respect to the two others. In the SIRANE model, these reactions are modelled assuming a photo-stationary equilibrium defined by the equation (2) where  $[\text{NO}]$ ,  $[\text{NO}_2]$ , and  $[\text{O}_3]$  refer respectively to the molar concentration of NO, NO<sub>2</sub>, and O<sub>3</sub>.

$$\frac{k_1}{k_3 [\text{O}_3]} = \frac{[\text{NO}]}{[\text{NO}_2]} \quad (2)$$

In the SIRANE model, the dispersion module is firstly implemented considering the emitted species are passive. After this dispersion step, the module relative to the chemistry is implemented. In the following paragraphs, the molar concentrations of the specie E before and after the implementation of the chemistry module are respectively noted  $[\text{E}^d]$  et  $[\text{E}]$ .

### Source apportionment

In this study, the source apportionment is implemented within the SIRANE model. The method consists on following tagged species emitted by the sources taken into account in the simulations. This approach is already used with regional scale dispersion models as CAMx (Wagstrom et al., 2008) and CMAQ (Kwok et al., 2013) but is still barely used at the urban scale, with an explicit representation of buildings. When the processes linking the emissions and the concentrations are linear, the estimation of the source contribution is quite trivial. The difficulty in the source apportionment is to deal with non-linear phenomena as chemical reactions. As Koo et al. (2009) said, the source apportionment can be estimated by several ways when chemical reactions are involved but there is not a reference method to which to compare.

The first method applied to deal with the chemical reaction assume that the initial (before chemistry) ratio  $[\text{NO}]/[\text{NO}_2]$  is larger than the photo-stationary ratio  $[\text{NO}]/[\text{NO}_2]$  (after chemistry). So,  $[\text{NO}]$  having to decrease and  $[\text{NO}_2]$  having to increase to reach the photo-stationary equilibrium, we consider that the Chapman cycle can be simplified by the reaction (3).



The equation (3) indicates that the concentration variation of NO, NO<sub>2</sub>, and O<sub>3</sub> can be estimated with the NO concentration variation. Therefore, the source contribution is estimated with the equation (4) where the subscript g indicates that the concentration is due to the contribution of the source g (source contribution). The equation (4) indicates that [E]<sub>g</sub> is always function of [NO<sup>d</sup>]<sub>g</sub>. Moreover, we can note that it is necessary to emit NO species to contribute to the NO concentrations. However, looking for the Chapman cycle, it is possible to contribute to NO concentrations emitting NO<sub>2</sub> species. Finally, it is important to notice that in this first approach, the source contribution can be negative when [E] is inferior to [E<sup>d</sup>].

$$[E]_g = [E^d]_g + ([E] - [E^d]) \frac{[NO^d]_g}{[NO^d]} \quad (4)$$

The second method proposed to estimate the source contribution for the chemical species assumes that the photo-stationary equilibrium is quickly reached. After having reached the equilibrium, we consider that the Chapman cycle can be resume as a dynamic equilibrium represented by the reaction (5).

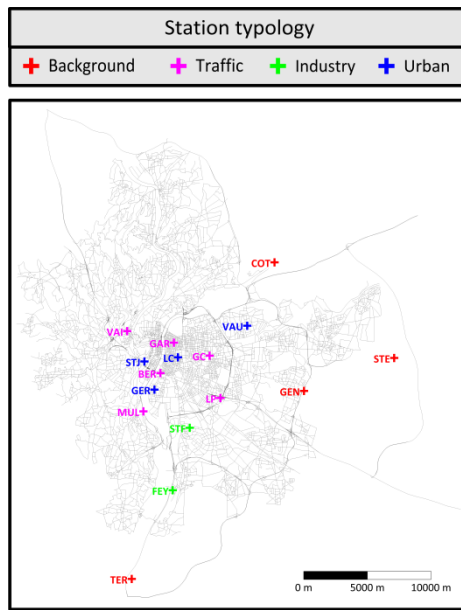


Moreover, we suppose in this method that the source contribution for the NO<sub>x</sub> species is similar before and after the chemical reaction and we consider that the reaction (5) leads to a source contribution similar (proportionally) for each species. So the source contribution with the second method is estimated with the equation (6) where the subscript g indicates that the concentration is due to the contribution of the source g (source contribution). With this method, the source contribution is always positive (or null). Moreover, the formulation of the equation (6) shows that sources emitting NO or NO<sub>2</sub> species contribute necessarily to the concentrations of the NO, NO<sub>2</sub>, and O<sub>3</sub> species.

$$[E]_g = [E] \frac{[NO^d]_g + [NO_2^d]_g}{[NO^d] + [NO_2^d]} \quad (6)$$

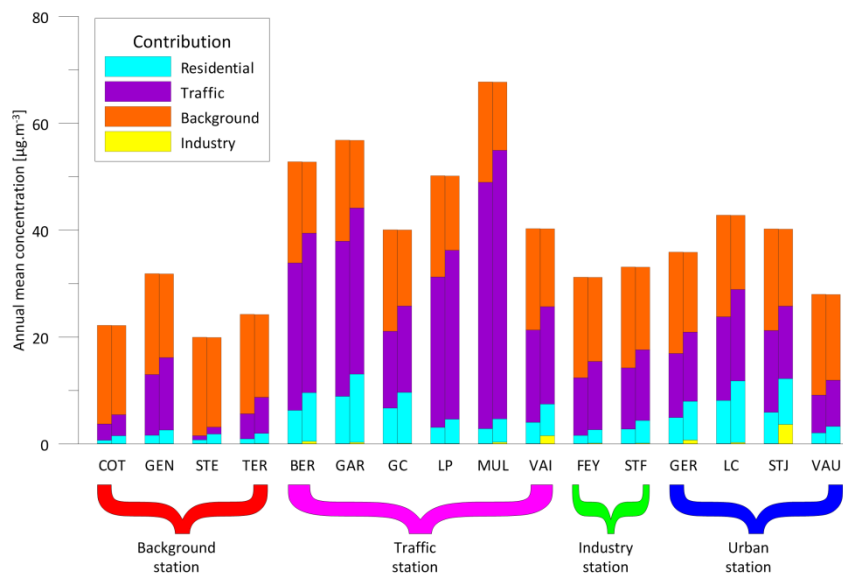
## Results

These two methods are applied with the SIRANE urban air quality model to estimate the industry, the tertiary, the traffic, and the background contributions to the NO<sub>2</sub> annual mean concentration in the Lyon city in 2008. The Figure 1 shows the domain study where 16 monitoring stations are available. These stations can be classified into four categories according to the environment in which they are located: the sub-urban stations (COT, GEN, STE, and TER), the traffic stations (BER, GAR, GC, LP, MUL, and VAI), the industrial stations (FEY and STF) and the urban stations (GER, LC, STJ, and VAU). The Figure 2 shows the source contribution estimated for the locations of these monitoring stations.



**Figure 1.** Study case domain and the localization of the monitoring stations

The results of the two methods are quite similar. In the two cases, the industry contribution is low compared to the three other sources. We can note that the industry contribution is only distinguishable with the second method. Moreover, we can remark that the background contribution is slightly lighter with the second method. The two methods lead to pretty consistent results for the traffic stations since the main contribution for these stations comes from the traffic. Likewise, the results are consistent for the sub-urban stations where the main contribution comes from the background concentration. We note for each method that the contribution profiles for the industry and urban stations are similar. This is due to industry contribution which is relatively light for the  $\text{NO}_2$  specie.



**Figure 2.** Source apportionment to  $\text{NO}_2$  concentrations determined with the 2<sup>nd</sup> model

## Conclusion

This study compares two methods implemented within the SIRANE urban air quality model to estimate the industry, the tertiary, the traffic, and the background contributions to the NO<sub>2</sub> annual mean concentration on the Lyon city in 2008. The approach consists on following the emitted species during the dispersion. The difference between the two methods concerns the treatment of the non-linear process induced by the chemical reactions which occurs in the Chapman cycle. The first method supposes that the source contribution can be estimated in function of the NO concentration variation whereas the second consists on estimating the source contribution in compared to the NO<sub>x</sub> concentration. The results for the two methods are pretty consistent since the majority contribution for the sub-urban and the traffic stations comes from the background concentration and the traffic emissions respectively.

- Bonyoung Koo, G.M.W., 2009. Comparison of Source Apportionment and Sensitivity Analysis in a Particulate Matter Air Quality Model. *Environ. Sci. Amp Technol.* 43, 6669–75. doi:10.1021/es9008129
- Kwok, R.H.F., Napelenok, S.L., Baker, K.R., 2013. Implementation and evaluation of PM<sub>2.5</sub> source contribution analysis in a photochemical model. *Atmos. Environ.* 80, 398–407. doi:10.1016/j.atmosenv.2013.08.017
- Seinfeld, J.H., 1986. *Atmospheric Chemistry and Physics of Air Pollution*, 1 edition. ed. Wiley-Interscience, New York.
- Soulhac, L., 2000. *Modélisation de la dispersion atmosphérique à l'intérieur de la canopée urbaine*. Ecully, Ecole centrale de Lyon.
- Soulhac, L., Salizzoni, P., Cierco, F.-X., Perkins, R., 2011. The model SIRANE for atmospheric urban pollutant dispersion; part I, presentation of the model. *Atmos. Environ.* 45, 7379–7395. doi:10.1016/j.atmosenv.2011.07.008
- Soulhac, L., Salizzoni, P., Mejean, P., Didier, D., Rios, I., 2012. The model SIRANE for atmospheric urban pollutant dispersion; PART II, validation of the model on a real case study. *Atmos. Environ.* 49, 320–337. doi:10.1016/j.atmosenv.2011.11.031
- Wagstrom, K.M., Pandis, S.N., Yarwood, G., Wilson, G.M., Morris, R.E., 2008. Development and application of a computationally efficient particulate matter apportionment algorithm in a three-dimensional chemical transport model. *Atmos. Environ.* 42, 5650–5659. doi:10.1016/j.atmosenv.2008.03.012
- WHO, 2014. WHO | 7 million premature deaths annually linked to air pollution [WWW Document]. WHO. URL <http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/> (accessed 10.8.15).