



## Measurement of particle number and related pollutant concentrations in an urban area in South Brazil



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### HIGHLIGHTS

- ▶ Particle number and its association with NO, NO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub> was analyzed.
- ▶ Results showed a higher number concentration of the PR2.5 and PR1.0 size ranges.
- ▶ Particle number concentration was influenced by emissions from motor vehicles.
- ▶ Correlation analysis suggest similar emission sources for NO, NO<sub>x</sub> and particle number.
- ▶ Results indicate influence of meteorological conditions on particle number.

### ARTICLE INFO

#### Article history:

Received 22 August 2012

Received in revised form

6 December 2012

Accepted 14 January 2013

#### Keywords:

Air quality

Particle number

Nitrogen oxides

Ozone

### ABSTRACT

The purpose of the present study was to analyze atmospheric particle number concentration at Sapucaia do Sul, in the Metropolitan Area of Porto Alegre, and associate it with the pollutants NO, NO<sub>2</sub>, and O<sub>3</sub>. Measurements were performed in two periods: August to October, in 2010 and 2011. We used the following equipment: the continuous particulate monitor (CPM), the chemiluminescent nitrogen oxide analyzer (AC32M), and the UV photometric ozone analyzer (O342M). Daily and hourly particle number concentrations in fractions PR1.0 (0.3–1.0 μm), PR2.5 (1.0–2.5 μm), and PR10 (2.5–10 μm), and concentrations of pollutants NO, NO<sub>2</sub>, NO<sub>x</sub>, and O<sub>3</sub> were measured. These data were correlated with meteorological parameters such as wind speed, temperature, relative humidity, and solar radiation. The daily variation of OX (NO<sub>2</sub> + O<sub>3</sub>) and its relation with NO<sub>2</sub> were also established. The results obtained for daily particle number concentration (particles L<sup>-1</sup>) showed that the area of study had higher particle number of PR2.5 and PR1.0 size ranges, with values of 19.5 and 28.51 particles L<sup>-1</sup>, respectively. Differences in particle number concentrations in PR1 and PR2.5 size ranges were found between weekdays and weekends. The daily variation per hour of concentrations of particle number, NO, and NO<sub>x</sub> showed peaks during increased traffic flow in the morning and in the evening. NO<sub>2</sub> showed peaks at different times, with the first peak (morning) 2 h after the peak of NO, and a second peak in the evening (19:00). This is due to the oxidation of NO and to the photolysis of NO<sub>3</sub> formed overnight. Correlation analysis suggests that there may be a relationship between the fine and ultrafine particles and NO, probably indicating that they have similar sources, such as vehicular emissions. In addition, a possible relationship of solar radiation with fine particle number concentrations, as well as with O<sub>3</sub> was also observed. The results, too, show an inverse relationship between particle number concentration and relative humidity.

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### 1. Introduction

Among many factors that generate air pollution, emission of pollutants from motor vehicles (Colville et al., 2001) is a major source of particulate matter (Bathmanabhan and Saragur, 2010) and NO (Gaffney and Marley, 2009) in urban areas. With regard to particles emitted by mobile sources, over 90% of the particle

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number concentration belong to the  $<1 \mu\text{m}$  fraction (Morawska et al., 2008). However, particle emission depends on temperature, with higher concentrations at low temperatures (Olivares et al., 2007), and higher sulfur content in fuel, e.g. in diesel (Wählin, 2009). Moreover, the addition of biodiesel to diesel contributes to a decrease in particle emission due to a lower formation of sulfate particles during combustion (Dwivedi et al., 2006).

Although automotive vehicles are the main source of fine particles in urban areas, the number and size distribution of particles can change rapidly due to the influence of transformation processes, such as coagulation and condensation, and to turbulence, which improves mixing and dilution (Kumar et al., 2011). Moreover, vehicle emissions are a major source of nitrogen oxides emissions, especially in the form of NO (Gaffney and Marley, 2009), causing an almost direct emission in the boundary layer. Diesel engines produce five times the amount of  $\text{NO}_x$  by mass of burnt fuel compared to gasoline vehicles. Furthermore, the addition of biodiesel to diesel can slightly increase  $\text{NO}_x$  emissions (Coronado et al., 2009).

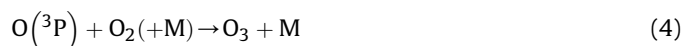
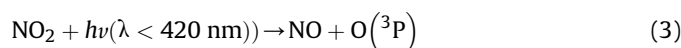
Although not having significant natural or anthropogenic emission sources (PORC, 1997), secondary pollutants such as  $\text{NO}_2$  and tropospheric  $\text{O}_3$  may be formed by different reactions of primary pollutants in the air (Finlayson-Pitts and Pitts, 2000). According to Jenkin and Clemitshaw (2000), the main conversion reactions between NO and  $\text{NO}_2$  are reactions (1), (2), (3), and (4):



However, under most tropospheric conditions, reaction (1) is not significant, and the major way by which NO is converted to  $\text{NO}_2$  is by reaction (2) with  $\text{O}_3$ :



Under the influence of solar radiation during the day,  $\text{NO}_2$  undergoes photolysis and reconverts to NO by reactions (3) and (4) below:



where M is a third compound (typically  $\text{N}_2$  or  $\text{O}_2$ ), which absorbs the vibrational energy in excess and thereby stabilizes the  $\text{O}_3$  molecule formed,  $h\nu$  is the energy of one photon ( $\lambda < 424 \text{ nm}$ ), and  $\text{O}({}^3\text{P})$  is active monatomic oxygen (Han et al., 2011). Photolysis of  $\text{NO}_2$  is an important source of  $\text{O}_3$  generation (Zielinska, 2005).  $\text{NO}_x$  has a short chemical lifetime and its major effects on  $\text{O}_3$  are limited to its proximity to emission sources (Uherek et al., 2010).

Tropospheric  $\text{O}_3$  is formed by several complex photochemical reactions between nitrogen oxides (NO and  $\text{NO}_2$ ) and volatile organic compounds (VOCs) in the presence of sunlight. However, in urban areas, concentrations of  $\text{O}_3$  are usually lower than in rural areas due to its reaction with NO (from vehicle emissions) to produce  $\text{NO}_2$ . Sunlight starts the process by supplying UV radiation that dissociates certain stable molecules, leading to the formation of hydrogen free radicals ( $\text{HO}_x$ ). In the presence of  $\text{NO}_x$ , these free radicals catalyze the oxidation of VOCs to form  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . Partially oxidized organic species are generated as intermediate oxidation products, such as  $\text{O}_3$  formed as a byproduct.  $\text{O}_3$  can be formed from the photolysis of  $\text{NO}_2$  by reactions (3) and (4). In the presence of NO, the degradation reactions of VOCs lead to the formation of intermediate  $\text{RO}_2$  and  $\text{HO}_2$  radicals. These radicals react with NO converting it to  $\text{NO}_2$  (Atkinson, 2000).

Within minutes, reactions (2) and (3) reach a balance and a so-called photostationary state, determined by Leighton (1961):

$$[\text{NO}] \cdot [\text{O}_3] / [\text{NO}_2] = J_1 / k_4 \quad (5)$$

Where  $J_1$  is the rate of  $\text{NO}_2$  photolysis, which varies during the day according to the solar zenith and radiation transmission through the atmosphere (Kewley and Post, 1978), and  $k_4$  is the reaction rate coefficient of NO with  $\text{O}_3$ , as a function of temperature (Jenkin and Clemitshaw, 2000; Notario et al., 2012) so that various experimental values may be found, depending on the temperature range in which the measurements were taken. The value of  $k_4$  in reaction (5) was calculated using the equation of Seinfeld and Pandis (2006):  $k_4 (\text{ppm}^{-1} \text{ min}^{-1}) = 3.23 \times 10^3 \exp[-1430/T]$ .

There are few studies on the atmospheric chemistry in the area of study and Brazil, specifically analyzing particle number concentrations in PR1 (0.3–1.0  $\mu\text{m}$ ), PR2.5 (2.5–1.0  $\mu\text{m}$ ), and PR10 (10–2.5  $\mu\text{m}$ ) size ranges, which are now being measured for the first time. This work is part of an attempt to investigate the particle size distribution of PR1, PR2.5, and PR10 size ranges and its relationship with gaseous pollutants ( $\text{O}_3$ , NO,  $\text{NO}_2$ , and  $\text{NO}_x$ ) and weather conditions. This is important because the study of particle size distribution allows a more accurate assessment of the source of the particulate matter formed in the area of study, as well as of NO,  $\text{NO}_2$ , and  $\text{O}_3$ . We also examined the variation of OX ( $\text{NO}_2 + \text{O}_3$ ) and the rate of  $\text{NO}_2$  photolysis, besides the differences in weekend and weekday concentrations.

## 2. Materials and methods

### 2.1. Area of study

The sampling site Sapucaia do Sul (Fig. 1) is located in the area of study: the Metropolitan Area of Porto Alegre (MAPA). The area of study is located in central-eastern part of the Brazilian state of Rio Grande do Sul. It is delimited by parallels  $28^\circ\text{S}$  and  $31^\circ\text{S}$  and longitudes  $50^\circ\text{W}$  and  $54^\circ\text{W}$ . The area currently comprises 9652.54  $\text{km}^2$  and, according to estimates by METROPLAN (the State Foundation for Metropolitan and Regional Planning), has 4,101,032 inhabitants (METROPLAN, 2012).

The region is the most urbanized area of the state and is characterized by different industries, including several stationary sources such as the Alberto Pasqualini Refinery, steel mills that do not use coke (Siderurgica Rio-grandense and Aços Finos Piratini), the III Petrochemical Industrial Complex, and coal-fired power plants (Termelétrica de Charqueadas – TERMOCHAR, and the Usina Termelétrica de São Jerônimo – USTJ). However, this site has low industrial influence upstream of the prevailing winds. It is estimated that the most significant contribution are mobile sources due to the great number of vehicles in the region, which represent 20% of the total cars of the state's fleet (Dallarosa et al., 2008). This region has a high vehicles traffic, approximately 1,840,703 vehicles (DETRAN, 2012), of which circa 84.98% is fueled by gasoline, 6.67% by diesel, 6.58% by ethanol, 1.03% by natural gas, and 0.74% has flex technology (Teixeira et al., 2008). The sampling site is also near the lower axis of the BR-116 highway, where the daily flow is approximately 150,000 vehicles (MT, 2012), and traffic jams and slow speeds are frequent. Currently, gasoline-fueled vehicles use a mixture of gasoline and 20% of ethanol (PETROBRAS, 2012). The diesel used in the MAPA has a level of 500 ppm of sulfur and it is added of 5% of biodiesel (Mattiuzzi et al., 2012). A study by Teixeira et al. (2012) shows that from the total emissions originating from mobile sources within the MAPA, diesel vehicles are responsible for most emissions of particulate matter and  $\text{NO}_x$ .

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