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Aerosol removal due to precipitation and wind forcings in Milan urban area

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ABSTRACT

Air pollution represents a critical issue in Milan urban area (Northern Italy). Here, the levels of fine particles increase, overcoming the legal limits, mostly in wintertime, due to favourable calm weather conditions and large heating and vehicular traffic emissions. The main goal of this work is to quantify the aerosol removal effect due to precipitation at the ground. At first, the scavenging coefficients have been calculated for aerosol particles with diameter between 0.25 and 3 μm. The average values of this coefficient vary between 2×10^{-5} and 5×10^{-5} s^{-1}. Then, the aerosol removal induced separately by precipitation and wind have been compared through the introduction of a removal index. As a matter of fact, while precipitation leads to a proper wet scavenging of the particles from the atmosphere, high wind speeds cause enhanced particle dispersion and dilution, that locally bring to a tangible decrease of aerosol particles' number. The removal triggered by these two forcings showed comparable average values, but different trends. The removal efficiency of precipitation lightly increases with the increase of particle diameters and vice versa happens with strong winds.

1. Introduction

Milan is the most industrialized and densely populated city of Northern Italy and is characterized by aerosol concentrations, which often exceed the limits set by the European air quality Directive (Commission and Directive, 2008). Its location, in the middle of the Po Valley, is characterized by the presence of the Alps to the North and West sides, and the Appennines to the South side, which form a natural barrier protecting the city and generally the valley from the major circulations coming from Northern Europe and from the sea. Consequently, rainfall events are limited (the average annual precipitation is less than 1000 mm) and winds are generally weak or absent. In Milan, the suburbs are in continuous demographic growth, the metropolitan area reaches about 6,000,000 residents (Silibello et al., 2008), causing an increase of vehicular traffic. In wintertime, the coupling of traffic and heating emissions together with calm weather and frequent thermal inversions worsen the air pollution conditions, making Milan to be one of the most polluted cities in all the European continent. However, some cities of the Po Valley (such as Cremona, Pavia or Turin) and also some industrial areas (such as the one in Brescia), can have aerosol concentrations comparable with the ones in Milan, due to the specific orographic and geomorphological characteristics of this region.

The high suspended aerosol concentrations in low atmosphere cause short-term health effects and increases the possibility of contracting serious acute or chronic respiratory and cardiovascular diseases (Pope, 2000). The reduction of visibility (Schwartz, 1996) and the modification of the local climate through the alteration of Earth's radiation balance (Finlayson-Pitts and Pitts, 2000) are two additional environmental issues related to this phenomenon. Under this context, it is important to understand what are the natural mechanisms which can decrease the aerosol particles' numbers and how they work.

Firstly, the precipitation plays an important role in aerosol removal through the below-cloud scavenging process, by which the falling particles (rain droplets and ice crystals) deposits aerosol particles on the ground surface. This process is influenced by rain droplet size distribution, rain intensity and collision efficiency between particles and rain droplets (Seinfeld and Pandis, 2006; Laakso et al., 2003). The latter is affected by two main phenomena: Brownian diffusion and inertial impaction. The aerosol particles in the atmosphere move according to a random motion, the Brownian diffusion, which decreases with the increasing of the particle size ($d_p < 0.2$ μm). Some of these particles collide with the falling rain-

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drops and are deposited on the ground. The deposition increases with the decrease of the particle diameter. Conversely, inertial impaction is a process characteristic of particles above a certain size (typically $d_p > 1$ μm), which, thanks to their inertia, are not able to follow the streamlines of the raindrops and thus easily impact the drops' surface. For these reasons, the scavenging efficiency reaches a minimum in the range between 0.2 and 1 μm, which is usually defined as the "Greenfield gap" (Greenfield, 1957). Particles in this range are too big to be affected by Brownian diffusion, but at the same time, they do not have enough mass to be exposed to inertial impaction.

Secondly, high wind speeds clean the atmosphere by dispersing and diluting the aerosol particles and preventing local accumulations (Hussein et al., 2006; Kgabi and Mokgwetsi, 2009; Hussein et al., 2011). The occurring of high wind speeds (5 m/s or greater) in a urban and air polluted area like Milan, where several aerosol anthropogenic sources are located, causes an average local decreasing of aerosol particles' numbers. For this reason, the wind dispersion can be viewed as a local removal process. What one can expect is a wind removal efficiency which is negatively correlated with the aerosol particle size (Harrison et al., 2001). The smallest and lightest particles (which are usually suspended for long periods in the atmosphere) can be easily dispersed and diluted by the action of high wind speed, causing a local decrease of the fine particles' concentration. The largest and heaviest particles (characterized by short suspension time) can be transported for shorter distances and can also be subjected to resuspension phenomena, due to turbulent air flows, caused by the blowing of strong winds. Under this process, the particles that were previously deposited on the surface of any substrate are resuspended, therefore it is possible to measure local and temporal increase of the coarse particles' concentration. It is important to clarify that these assumptions are true when the study area is far away from the sea. In this case, high wind speed can bring to an increase of the aerosol numeric concentration because of sea-salt particles production at the ocean surface by bursting of air bubbles resulting from the entrainment of air induced by wind stress (Pant et al., 2008).

There are different studies (Davenport and Peters, 1978; Nicholson et al., 1991; Volken and Schumann, 1993; Mircea et al., 2000; Laakso et al., 2003; Chate and Pranesha, 2004; Maria and Russell, 2005; Andronache et al., 2006; Zikova and Zdimal, 2016) where precipitation scavenging coefficients have been calculated for different size ranges and usually compared to theoretical predictions. In almost all these works, the analyses have been performed in sub-rural sites or in areas characterized by an overall aerosol load lower than the one in Milan. Here, we calculated the rain scavenging coefficient within Milan urban area for aerosol particles in 0.25–3 μm size interval and we compared the results with the studies of Davenport and Peters (1978) and of Volken and Schumann (1993), analysing any similarity and dissimilarity. Furthermore, we compared the removal efficiency of precipitation and winds through the introduction of a removal index, in order to clarify how these two processes act. The paper is organized as follows: a summary of the theory of the aerosol removal mechanisms is reported in Section 2; the instruments, the data acquisition and processing are presented in Section 3; the results are discussed in Section 4; the conclusions are given in the last section.

2. Theory of the mechanisms of aerosol removal

During a precipitation event, some of the precipitation particles falling through the air collide with the aerosol particles, collecting and taking them at the ground. This process is called precipitation below-cloud scavenging. The temporal variability of the aerosol concentration due to this mechanism is described by Eq. (1) (Seinfeld and Pandis, 2006):

$$\frac{\mathrm{d}c(d_p)}{\mathrm{d}t} = -\lambda c(d_p) \tag{1}$$

where, c is the concentration of the aerosol particles with equivalent diameter d_p [μm] and λ [1/s] is the scavenging coefficient due to precipitation collection. Many factors influence this coefficient: the size of the aerosol particle; the size, the velocity and the concentration of the precipitation particles; the collection efficiency of precipitation. Since all of these are usually unknown, it is possible to indirectly calculate the scavenging coefficient (assuming that rain collection is the only aerosol removal mechanism), by integrating Eq. (1) in time (Sperber and Hameed, 1986; Laakso et al., 2003):

$$\lambda(d_p) = \frac{1}{t_1 - t_0} \ln\left(\frac{c_0(d_p)}{c_1(d_p)}\right) \tag{2}$$

In Eq. (2), all the terms involved are known: c_0 and c_1 are the aerosol concentrations at the beginning (t_0) and at the end (t_1) of the rain event, while ($t_1 - t_0$) is the event duration in seconds. When precipitation scavenging cannot be considered the unique mechanism influencing the aerosol dynamics, Eq. 1 should be written as:

$$\begin{aligned}\frac{\mathrm{d}c(d_p)}{\mathrm{d}t} = &-\lambda c(d_p) \pm \left[\frac{\mathrm{d}c(d_p)}{\mathrm{d}t}\right]_{advection} \pm \\ &\left[\frac{\mathrm{d}c(d_p)}{\mathrm{d}t}\right]_{hygr.\,growth} \pm \left[\frac{\mathrm{d}c(d_p)}{\mathrm{d}t}\right]_{turb.\,mixing} \pm \\ &\left[\frac{\mathrm{d}c(d_p)}{\mathrm{d}t}\right]_{instr.\,errors} + \left[\frac{\mathrm{d}c(d_p)}{\mathrm{d}t}\right]_{emissions} \pm \ldots\end{aligned} \tag{3}$$

Eq. (3) shows some of the most important mechanisms (advection, hygroscopic growth, turbulent mixing, instrumental errors, aerosol emissions), whose influences cannot be easily ignored when one wants to investigate the aerosol dynamics. In some cases, these contributions can indeed be greater than the effect of precipitation collection. Conversely, if there are no other contributions except precipitation removal, all the right terms of Eq. (3), except the precipitation collection, can be neglected (Laakso et al., 2003).

For example, the air mass changes due to horizontal and vertical advection can cause errors in the evaluation of the precipitation scavenging efficiency. This effect is low when stratiform winter events are taken into account. High relative humidity causes the hygroscopic growth of small particles, leading to an increase of coarse particles and to a decrease of fine particles. Turbulent mixing can bring to space and temporal changes of aerosol concentration by mixing two different air masses, with different aerosol levels. The instrumental errors are often due to wrong calibrations of the instruments or discrepancies caused by the different location of the instruments (in case rain and aerosol concentration are not measured in the same exact site). Furthermore, when there is a very small number of aerosol particles, high fluctuations in the collection scavenging efficiency can be found. For example, in a urban and very polluted site as Milan, the number of particles greater than 3 μm is heavy less than 100, and the correspondent value of the scavenging coefficient is characterized by a low level of significance. Besides, it is important not to forget the influence of the numerous punctual aerosol emissions, features of an urban site. In particular the vehicular traffic is characterized by a recursive trend consisting of two positive peaks (corresponding to work/school entrance and exit), which lead to an increment of the aerosol concentration.

In order to determine the scavenging coefficient, the contributions of the non-collection terms have to be considered. Given the difficulties in the determination of the former contributions,