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Particulate matter characterization in a steelworks using conventional sampling and innovative lidar observations

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ABSTRACT

Particulate matter (PM) emissions from steelworks cause public concern. Although end-of-pipe and process integrated measures have led to a significant drop in emissions of large particles from stacks, fine aerosols were not specifically considered, nor were emissions from fugitive and open sources. In this study, we deployed aerosol samplers together with a scanning ultra-violet (UV) lidar to characterize total suspended particles (TSP), PM_{10}, and $PM_{2.5}$, in emissions from a large integrated steelworks in Spain over a 16-day period. We determined the content of carbonaceous, soluble inorganic, mineral dust, and metal species. A positive matrix factorization was carried out on our dataset. Despite mineral dust being predominant in all size fractions, the steelworks was clearly a source of carbonaceous species, resulting in production of secondary inorganic aerosols. In particular, stack emissions were a major contributor of fine particles, while open sources dominated the emissions of TSP, yielding up to 80% of particles larger than PM_{10}. UV lidar provided 2D maps of aerosols in real time, with an ability to detect PM emissions and to visualize complex plumes. We suggest that air quality management of steelworks needs to focus on controlling large and coarse particle emissions, especially those from open sources.

Introduction

Particulate matter (PM) emissions have always been an environmental challenge for steel makers. For logistical reasons, steel factories are often located close to urban areas and are consequently subjected to complaints about PM-emissions from the public. While total suspended particles (TSP) have long been the only target, standards for PM_{10} (particulate material, with an aerodynamic diameter below 10 μm) have been implemented since the beginning of the 2000s in most steelworks in industrialized countries. In Europe, abatement technologies were adopted, in agreement with best available techniques (BAT), as described in the BAT reference documents (BREFs) from the European Union (EU) under the Integrated Pollution Prevention and Control Directive (EU Directive No. 96/61/CE). Among these BREFs, those relating to iron and steel production, as well as storage of bulk or dangerous materials are the most extensively adopted (EIPPCB, 2006).

Emissions from steelworks are categorized as stack emissions, fugitive emissions (from industrial processes, but not released through stacks), and emissions from open sources (not from industrial processes). The installation of BAT has led to a significant drop in emissions of large particles in the atmosphere through the application of end-of-pipe and process integrated measures. Those measures also succeeded in decreasing several other pollutants along with PM. However, the contributions of open sources and the emissions of fine PM have not yet been considered.

The number of studies investigating the chemical characterization of aerosols inside or in the vicinity of steelworks has significantly increased over the last 5 years. In particular, several source apportionment studies have been published based on non-metallic datasets (Dall'Osto, Drewnick, Fisher, & Harrison, 2012) or on highly time-resolved observations of trace metal contents in aerosols (Choël, Deboudt, Flament, Aimoz, & Mériaux, 2007; Choël, Deboudt, & Flament, 2010; Park & Dam, 2010; Hleis et al., 2013). As one would expect, the resulting source profiles were defined by Fe-rich components, together with Zn, Mn, and Cr oxides. Some authors also noted that source profiles could markedly vary within the same industrial installation, depending on their processes (Taiwo et al., 2014a), the type of background aerosols, or setting, e.g., urban, rural, and/or marine (Dall'Osto, Booth, Smith, Fisher, & Harrison, 2008; Taiwo, Beddows, Shi, & Harrison, 2014b). It is interesting to note that most of these published studies were performed close to a steelworks located in Port Talbot, South Wales;

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although others took place in Dunkirk, a major industrial harbor in the North of France, as well as in Northern Europe and the United States. None were carried out in a developing country, where not only the chemistry of the aerosol mix greatly differs, but also the control policies on emissions, especially when diffuse and open sources are taken into account.

Aerosol sizes are typically discriminated into two main aerosol size fractions: coarse aerosols having aerodynamic diameters between 2.5 and 10 μm; and fine aerosols having aerodynamic diameters below 2.5 μm. Studies of sub-micron sized aerosols have been focused on the presence of Fe and Mn core–shells (Konarski, Iwanejko, Mierzejewska, & Diduszko, 2001), specific source apportionments (Choël et al., 2007, 2010; Taiwo et al., 2014a, b), and their hygroscopic characteristics (Sorooshian et al., 2012). Health effects of industrial aerosols are of course a key motivation to many such studies, especially with regard to the finest particles in the atmosphere (Strak et al., 2012), either from an occupational viewpoint (Górny, Szponar, Larsson, Pehrson, Prażmo, & Dutkiewicz, 2004) or as a public health concern (Oravisjärvi, Rautio, Ruuskanen, Tiittanen, & Timonen, 2008).

In this context, one could surmise that steelworks should focus on reducing their fine particle emissions as their main new target. However, based on observations in Port Talbot, Hayes and Chatterton (2009) concluded that steelworks appear to be a more significant source of coarse particles than of fine ones. To verify this assertion, we collected data for TSP, PM_{10}, and $PM_{2.5}$ on site, while air quality data were obtained from the local environmental protection agency, supplemented by observations obtained from a scanning ultra-violet (UV) lidar. We considered mass concentration together with chemical and trace metal composition for these different size fractions in various types of emissions from the steelworks. These data were used to determine particulate emissions on site in real time. Our results bring additional knowledge to the field of industrial emissions by examining the relative contribution of the main sources of particles, both inside and outside the plant, taking into account mass and chemical characteristics of three different size fractions.

Material and methods

Sampling site and meteorological conditions

A 3-week field campaign was carried out from 26 September to 14 October 2011. Our sampling site was located 10 m above ground level in the center of an integrated steelworks to facilitate the collection of aerosols originating from the main installations as well as from the regional background. The steelworks is located a few kilometers from Gijón, a harbor city of about 300,000 inhabitants in an agricultural and hilly area, but less than 2 km south of a coal-fired plant. The main processes taking place on site were iron-making (sintering, using a blast furnace, and raw materials dumping) and steel-making (using a basic oxygen furnace and coking). Fig. 1 shows a schematic layout of the steelworks, consisting of a complex mixture of stack sources (sintering plant, blast furnaces) and fugitive emissions (coke oven plant, blast furnace roof) associated with these main processes. Open source emissions, including wind dispersion of stockpiles of iron ore, blending processes, charging, transportation, and discharge of raw materials by conveyors or unsheeted tippers, also affect the aerosol characteristics of the plant.

The survey period can be divided into two distinct meteorological periods: a first half marked by variable conditions and low wind speed; and a second one marked by moderate to strong easterly winds. Rainfall only occurred on 6 and 7 October. Rain was associated with northerly winds, lower temperatures, and an increase in relative humidity (RH), surface pressure, and mixing layer height. Wind predominantly originated from the east (E; 47% of the time). Calms occurred 8% of the time, while slow winds were generally from the north (N) and west (W). Moderate to strong breezes mostly blew from the E and to a lesser extent from the NW. The occurrences of calms, E, SE, and N winds accounted for 95% of the total conditions during the field campaign, hence westerly winds will not be mentioned hereafter. The three main wind sectors are superimposed over the steelworks layout shown in Fig. 1. The average relative humidity remained above 60% most of the time. All meteorological parameters were monitored using an Oregon Scientific™ Professional Weather Center (mode WMR200A).

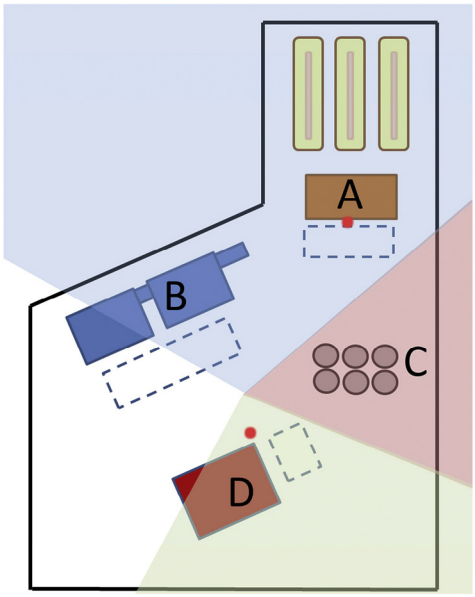

Fig. 1. Site layout indicating the location of all main aerosol sources: (A) sintering; (B) blast furnaces; (C) coking plant; and (D) foundry. The three main wind sectors are represented in blue (north, N); red (east, E) and green (southeast, SE). Sampling was carried out at the point where these three sectors meet. Lidar installation sites are shown as red dots. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

In situ aerosol sampling

PM_{10} and $PM_{2.5}$ samples were collected for a period of 16 consecutive days (28/09–13/10/2011). Every day at 8 am, two quartz fiber filters (Whatman, UK) and two polycarbonate Nuclepore filters (Whatman) were used to sample PM_{10} and $PM_{2.5}$ using URG 1-m^3/h PM cyclones (URG Corp., NC, USA). In addition, every 2 days, TSP were sampled on open-face Teflon filters (Whatman, UK) for trace metal analysis. In total, 32 quartz fiber filters and 32 polycarbonate filters were collected for analysis of carbonaceous and inorganic species, while eight Teflon filters were sampled for trace metal characterization. Quartz fiber filters were pre-cleaned at 800 °C for 24 h prior to sampling. Polycarbonate and Teflon filters were weighed before and after sampling, using a Mettler Microbalance with 1 μg readability, after equilibrating for 24 h at room temperature (23 ± 3 °C) under controlled relative humidity (<20%). The uncertainty in the gravimetric measurement was typically of the order of 10 μg, representing an average error of about 2%–3%.

Chemical analyses

Carbon analysis

Quartz filters were exposed to HCl fumes to remove carbonates. The total carbonaceous fraction of aerosols (TC), excluding

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