SESONAL AND VERTICAL VARIATION OF MINERALOGY AND PARTICLE SIZE DISTRIBUTION OF SETTLED DUST ALONG A HIGH BUILDING

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Kivonat

Az ülepedő porok jelentős hatással lehetnek a levegőminőségre, amelyben szemcseméretüknek és ásványos összetételüknek komoly szerepe van. Budapest egyik legforgalmasabb útja mentén álló toronyépület két oldalán, négy különböző magasságban, évszakonként gyűjtött ülepedő porok ásványos- és szemcseösszetételét vizsgáltuk. A minták szemcseeloszlására két csúcs jellemző (12 és 37 μm körül). A finomabb szemcsék aránya, de a maximális szemcseméret is a szélfelőli oldalon nagyobb. Jellegzetes vertikális eloszlási trend nem fedezhető fel, viszont nyáron a durvább szemcsék arányának növekedése jellemző. A fő ásványos alkotók a mintázási hely környezetének földtani jellemzőit tükrözik. A finom szemcsefrakcióban (<20 μm) az agyagásványok és az amorf anyag aránya jelentősen nő, és megjelenik az épület anyagából származtatható gipsz is. Az épület két oldala közti legnagyobb különbség a dolomit mennyiségében mutatkozott, ami az épület mögötti hegyről érkezik, és a speciális légáramlási viszonyoknak köszönhetően az utcafronton mutat nagyobb értékeket. A karbonát- és agyagásványok aránya nyáron jelentősen nő a kvarc rovására, ami a száraz felszínnel jellemzett időszakokat uraló porfelkavarodásnak tudható be. Az ásványos összetétel vertikális változása az agyagásványok esetén a legszembetűnőbb, és jól mutatja a kis szemcseméretű fázisokra jellemző kettős, helyi és távolabbi forrásokból is származó ülepedés egymással ellentétes hatását.

Abstract

Settled dust may affect air quality due to its mineralogical and particle size composition. We studied the mineralogical and particle size composition of settled dust collected seasonally at four heights and both sides of a high building next to a busy road in Budapest, Hungary. Particle size distribution of samples shows two maxima (at 12 and 37 μm). The ratio of fine particles and also the maximum particle size is higher at the windward side of the building. No characteristic vertical distribution pattern was observed. However, summer samples exhibit higher ratio of coarse particles. Major mineral dust components are characteristic of surrounding geology. Clay minerals and amorphous materials enrich in the fine particle fraction (<20 μm). Gypsum from constructing materials also appears there. Despite that dolomite is originated from dolomite hill behind the building its amount is higher at the front side due to special air flow conditions. In summer, carbonates and clay minerals show significant increase probably due to the re-suspension of local materials during the dry-surface periods. Vertical variation of dust mineralogy is the most conspicuous for clay minerals demonstrating the dual source of these phases as local and remote sources exert antagonistic effect on their deposition.

Introduction

Airborne particulate matter can be divided into the settled dust sediment and the suspended particulate matter. The former is composed of particles with great sedimentation power and their delay time in the atmosphere is very short. As causing generally near-source pollution, it is of important significance as an indicator of local air quality. Depending on their size and composition, such particles in the air may cause health disorders and may contaminate the soils and/or groundwater after their sedimentation.

In environments where population and traffic density are relatively high, the harmful effects of airborne particulate matter is expected to be significantly increased. This is often the case near busy traffic axis where urban topology may contribute to the creation of poor air dispersion conditions. Despite that its horizontal distribution characteristics is widely studied in the urban environment, there are no data about the deposition characteristics of major dust components at the heights typical for the urban environment. The aims of this study were 1) to investigate the distribution pattern of the particle size and mineralogical composition of the settled dust up to 33 metres height and 2) to study the seasonal variance for these parameters next to a busy road in Budapest, Hungary.

Materials and Methods

Bulk settled dust samples were collected seasonally for two years according to the Hungarian standard (MSZ 21454/1-83, 1983) using glass pots of 2000 mL containing 500 mL distilled water and 0.500 g of algaecide (analytical grade methyl 4-hydroxybenzoate) with continuous supply of the water. Altogether 8 sampling points were placed on the front and back sides of a building at a busy road at heights of 2, 9, 21 and 33 metres. The dust particles and water were separated by vacuum filtering. After weighing the dust, the samples were separated from the filters in ultrasonic bath. Selected samples were fractionated by wet sieving (with the mash size of 20 μm) to study the mineralogy of fine particle fraction.

The particle size distribution of the samples was studied by a Fritsch Analysette Microtech A22 laser diffraction analyser. Bulk mineralogical composition of the samples were analysed by a Philips PW 1710 X-ray diffractometer (XRD). The semi-quantitative phase analysis was carried out after Bárdossy et al. (1980). To characterize the phases below the detection limit of XRD, transmission electron microscopy (TEM) analyses were carried out on selected samples by a Philips CM20 instrument equipped with a Noran energy dispersive spectrometer (EDS). We pretended to analyse only one discrete particle in each case, which could be confirmed from the corresponding diffraction pattern.

Results and Discussion

Dust deposition exceeds the threshold limit value only occasionally with nearly threefold values in spring and summer (on average 9-10 g/m^2) than in other seasons. These differences can be due to the seasonal weather characteristics. Vertical dust deposition pattern mostly corresponds to the generally accepted air-flow model for tower buildings, except in summer when the local morphology of the natural and built environment and the weather characteristics together results in a specific distribution pattern (see in more details in Sipos and May, 2013).

The particle size distribution of the samples can be characterized generally by two maxima: one at around 37 μm and another one at around 12 μm. Additionally, a slight shoulder appears at around 2 μm forming a small peak for the 25% of the samples (Figure 1). This latter maximum is not characteristic of any season or sampling height. Between 17 and 61% of the samples is built up from particles smaller than 10 μm, so the ratio of inhalable particles may be seriously high (Zhao et al., 2010). In this case, there are no specific spatial characteristics for the individual sample pairs. On the contrary, the ratio of particles below 10 μm is lower in summer and spring than in winter and autumn. This is in accordance to the finding that the ratio of large particles is higher (the peak at 37 μm higher) in summer and spring than in winter and autumn. The average particle size distribution curves of the different sampling heights show, however, very similar pattern. The only differences are the unexpectedly low peak at 12 μm and the high one at 37 μm at 2 m and 33 m, respectively. The two maxima at 12 and 2 μm are higher by 10% at the back side of the building (windward side), e.g. the ratio of finer particles is higher there than at the front side (leeward side). On the contrary, the maximum particle size is slightly higher at the back (113 μm) than at the front side (90 μm).

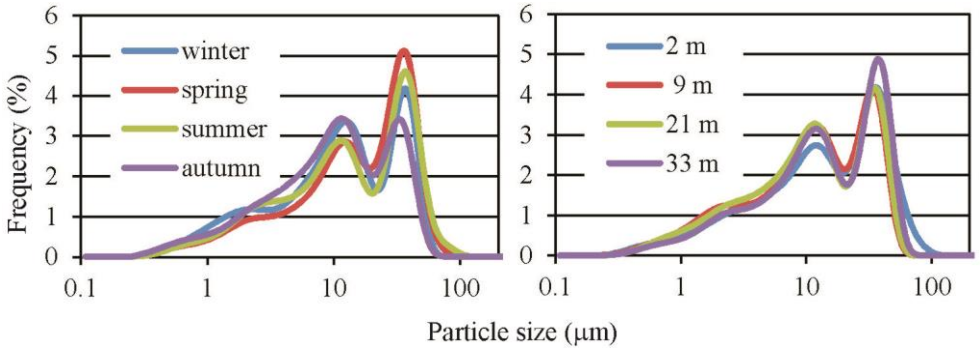

Figure 1. *Average particle size distribution curves for the different sampling heights and periods.*

The most frequent dust components are quartz (60-90 wt%), dolomite (0-20 wt%), calcite (0-15 wt%), feldspar (5-10 wt%), illite (1-5 wt%), chlorite (1-5 wt% and occasionally smectite (1-5 wt%). These phases are characteristic of surrounding geology and are the most common natural components of airborne particulate matter (Farkas and Weiszburg, 2006) although both silicates and carbonates may also originate from anthropogenic sources (Zajzon et al., 2013). In the fine particle size fraction (< 20 μm), the ratio of clay minerals and amorphous materials significantly increases and gypsum also appears originating probably from the construction materials and/or the reaction of sulphuric acid and calcic material in the air (Panigrahy et al., 2003).

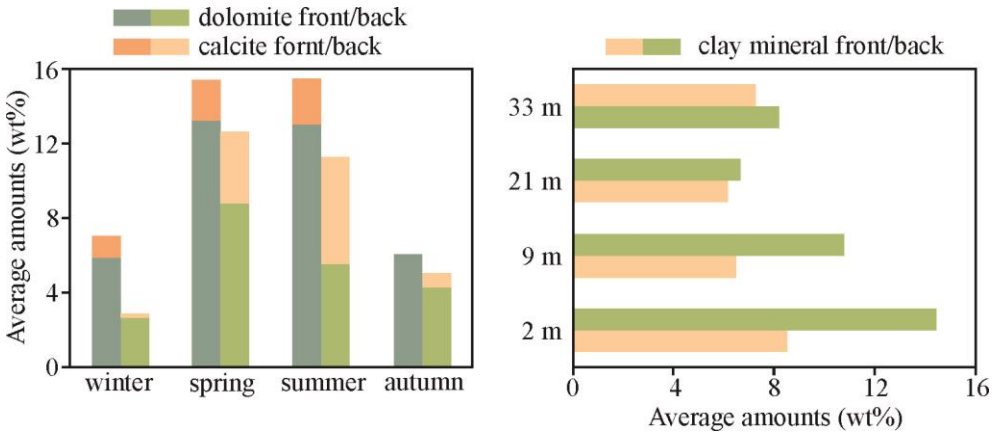

Figure 2. *Seasonal changes in the average amounts of carbonate minerals (left) and vertical changes in the average amounts of clay minerals (right) at the two sides of the building in the settled dust samples.*

The most spectacular spatiotemporal variation was found for the carbonate minerals (Figure 2). The average amount of dolomite may be even twice as much on the front as at the back side for each season despite the presence of the dolomite hill behind the building. The amount of calcite exceeds that of dolomite only at the back side of the building occasionally. Evidently, the Triassic dolomite rock is the source of dolomite. Due to the vicinity and morphology of the dolomite hill and the building, formation of a wake interface flow could be expected between them resulting in the transport and deposition of the hill's material on the leeward (front) side of the building in an isolated separation bubble (Oke, 1988). The amount of carbonate minerals is twice as much in the periods of large dust deposition as in winter and autumn. Clay minerals show similar seasonal behaviour to carbonates. This suggests their significant local source by re-suspension of street dust and local soil. However, clay minerals are the only phases showing characteristic vertical

differences in their deposition (Figure 2): their amount decreases with increasing sampling height at the back and much more uniform pattern was found at the front side of the building. The spatiotemporal variation in the clay mineral's deposition suggests the presence of remote sources additionally to local ones and these sources may have antagonistic effects at the different sides and levels of the building. Clay minerals are well known phases travelling very long distances (thousands of kilometres) in the air (Caqiuneau et al., 1998).

The major mineral constituents of the settled dust samples identified by XRD were also observed by our TEM analyses. Besides them, magnetite was found as a frequent component of the samples (Figure 3). This mineral often forms aggregates consisting of nano-sized (up to 100 nm) magnetite particles which may be attached to large silicate particles. On the other hand, magnetite is also present as relatively large (up to a few μm) spherular or xenomorphic polycrystalline particles. Both its size and morphology suggest that this mineral originates primarily from anthropogenic sources (Zajzon et al., 2013). Additional iron minerals, such as hematite and ilmenite were also found by TEM analyses but in much less amount than magnetite.

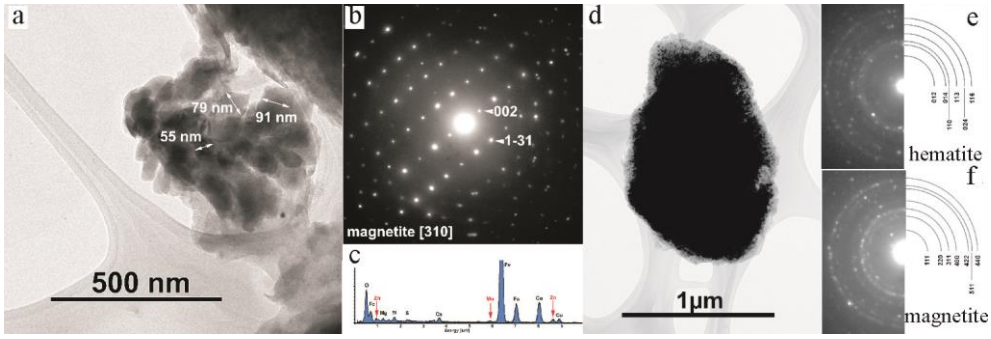

Figure 3. *TEM microphotograph of an aggregate consisting of magnetite single crystals (a), its diffraction pattern (b) and EDS spectra (c), as well as a bright field image of a polycrystalline aggregate of hematite and magnetite (d) and their diffraction patterns (e and f, respectively).*

Magnetite was also identified as the only magnetic phase in the these samples by magnetic analyses (Márton et al., 2011). These results suggested that magnetite particles are present mostly as fine superparamegnetic (<30 nm) particles on the one hand, and that the magnetic susceptibility of the samples (representing its magnetite content) show similar spatiotemporal characteristics to that of dust deposition. Additionally, the decrease of susceptibility with high frequencies of the settled dust samples was found to be very similar to that of vehicle exhaust material indicating that the primary source of this mineral is supposed to be the traffic.

Conclusions

Fine dust particles harmful to human health show seasonal settling characteristics and they enrich at sites prevented by local morphology. Not only the local dust components but also those originating from remote sources can be characterized by specific spatiotemporal deposition patterns.

Acknowledgements

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