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Glass-particle adhesion-force-distribution on clean (laboratory) and contaminated (outdoor) surfaces

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ABSTRACT

This paper explores whether laboratory-clean samples are representative of real outdoor surfaces – often covered in dust and other contaminants – with respect to adhesion properties, which ultimately affect particle resuspension. In this study, Atomic Force Microscopy (AFM) was used to measure the adhesion force between a clean, smooth glass particle (5 μm in diameter) and two glass slides, one prepared according to common laboratory practices (hereafter, clean glass) and a second one left outdoors for six weeks (hereafter, contaminated glass). It was found that the adhesion force distribution was bimodal for the clean glass while it showed three peaks for the contaminated glass. For both surfaces, the adhesion force showed a peak at very low adhesion values (< 100 nN) representing 1.2% of the measurements for the clean glass and 58% for the contaminated glass. This low-adhesion peak is important because it represents the particles that would be resuspended first even in light winds. The low adhesion-force measurements were associated with the presence of contaminants on the surface. Another notable difference between the adhesion force distributions was a long tail at high adhesion forces for clean glass (up to ~ 1900 nN) and the absence of the tail for the contaminated glass, which had a maximum adhesion force of 950 nN. The mean adhesion force on the contaminated glass was also lower than on the clean glass (206 ± 240 nN vs. 516 ± 354 nN). These observations indicate that the adhesion force distribution between a 5- μm smooth and clean glass sphere and the outdoor contaminated glass surface is quite different from that of the clean glass surface. Hence, one should be cautious when using adhesion force measurements from clean surfaces to estimate particle resuspension from outdoor surfaces.

1. Introduction

Airborne particles below 10 μm represent a health hazard because they can be inhaled and travel deep into the lungs (EPA, 2010). In urban environments, these particles can be either naturally-occurring (e.g., dust, pollen, and spores) or have an anthropogenic origin (e.g., soot, photochemical aerosols, radiological particles from an accident, biological particles from a terrorist attack). Once in the atmosphere, some of these particles will deposit on roads, buildings, vehicles, vegetation, and other urban surfaces. Wind-driven particle resuspension can bring some of these particles back into the atmosphere, resulting in a prolonged low-level secondary health hazard. In addition, for radiological and biological particles, their resuspension would result in decontamination and evacuation zones that change with time. It is therefore important to be able to estimate the fraction of deposited particles that will resuspend in

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specific weather conditions to safeguard the public's health.

There are a number of particle resuspension models in the literature that can assist in this task, e.g., [Reeks and Hall \(2001\)](#), [Zhang, Reeks, and Kissane \(2013\)](#), [Guingo and Minier \(2008\)](#), and [Henry and Minier \(2014a\)](#). These models estimate resuspension from the knowledge of the fluid dynamics around the particle (i.e., wind speed, turbulence intensity, aerodynamic forces) and the knowledge of the particle/surface interactions (i.e., the adhesion force). General reviews of aerodynamic and interfacial forces between a surface and a colloid particle can be found in [Ziskind \(2006\)](#), [Israelachvili \(2011\)](#), and [Henry and Minier \(2014b\)](#); for non-spherical particles, a broad discussion can be found in [Brambilla, Speckart, and Brown \(2017\)](#). The focus of this paper is on adhesion, therefore, only contributions related to this aspect will be discussed here. For micron-sized particles on a surface in air, the adhesion force can be seen as the sum of three components: the capillary force, the electrostatic force, and the van der Waals force ([Israelachvili, 2011](#)).

The capillary force is the result of the condensation of water between hydrophilic materials and for smooth surfaces, it is negligible below 30% relative humidity (RH) ([Wang, Qian, & Gao, 2009](#)); hence, it will not be discussed any further here because the humidity was below this value during the experiments described in this paper.

The electrostatic force in air is determined by the distribution of the charges on the particle and the surface ([Hubbard, Brockmann, Rivera, & Moore, 2012](#)). If adhesion is measured via Atomic Force Microscopy (AFM) like in this study, the presence of an electrostatic force can be inferred from the force vs. distance curves ([Jones, Pollock, Cleaver, & Hodges, 2002](#)). In our case, this force was absent and will also not be discussed any further here.

The van der Waals force is defined as the residual attractive or repulsive force between molecules or bodies that does not arise from a covalent bond or an electrostatic interaction. In this context, the van der Waals force is associated only with the weak London dispersion forces, which are almost always attractive. This force acts over a short range and decays rapidly to zero when two surfaces are separated. For ideally smooth surfaces and particles, it is proportional to the particle diameter (e.g., [Hamaker, 1937](#)) and existing theories correlate the adhesion force to surface deformation (e.g., [Johnson, Kendall, & Roberts, 1971](#); [Derjaguin, Muller, & Toporov, 1975](#)). Roughness on either the particle or the surface is known to reduce the adhesion force by reducing the contact area ([Fuller & Tabor, 1975](#)). One of the simplest models for predicting adhesion in presence of roughness was developed by [Rabinovich, Adler, Ata, Singh, and Moudgil, \(2000a, 2000b\)](#). This model is based on the concept that two scales of roughness can be used to describe the surface while the particle is considered perfectly smooth. The advantage of this model is that it requires only the Hamaker constant and the roughness statistics; the disadvantage is that it can only predict the average adhesion force instead of its distribution. [Kumar, Staedler, and Jiang \(2013\)](#) and [Laitinen, Bauer, Niinimäki, and Peuker \(2013\)](#) found that this model underestimates the mean adhesion force. [Rabinovich et al.'s](#) approach was extended to rod-shaped particles (like spores) in [Brambilla et al. \(2017\)](#). [Prokopovich and Perni \(2010\)](#) proposed a different approach based on a multi-asperity contact model derived from the idea that a number of surface asperities will come in contact with the particle. [Cooper, Gupta, and Beaudoin \(2001\)](#) pursued another approach in which the particle and surface are discretized into cylindrical elements and the adhesion force is computed as the sum of the pair-wise interactions. A similar approach was followed by [Jaiswal and Beaudoin \(2012\)](#). The accuracy of the last two models depends on the number of elements used to discretize the surface. Both the multi-asperity models and the discretization models provide a distribution of adhesion forces instead of just the mean value and require high-resolution roughness measurements.

There are numerous experimental studies on the van der Waals adhesion force, but most consider laboratory-clean particles and surfaces for reproducibility. However, our interest is in particle resuspension from outdoor urban surfaces, which are usually covered in dust, pollen, and other pollutants. This fact raises an important question: are the measurements taken on clean surfaces representative of real outdoor urban surfaces, i.e., are they valid surrogates? Intuitively, the adhesion force distribution on clean and dirty surfaces will not be the same, but here we are interested in quantifying the differences in the mean adhesion force and in comparing the shape of the distributions, as both quantities affect particle resuspension. The first goal of this work is therefore to investigate this question through establishing a baseline of adhesion properties for clean and contaminated surfaces.

It is worth noting that often resuspension models assume that the adhesion force can be described with a log-normal, a Gaussian or a Weibull distribution (e.g., [Reeks & Hall, 2001](#); [You & Wan, 2017](#)). However, measured distributions have sometimes been better approximated with other distribution functions and were bimodal at times (e.g., [Lyne, Wallqvist, & Birgisson, 2013](#)). Therefore, the second goal of this paper is to investigate the shape of the adhesion force distribution to determine if a resuspension model like [Reeks and Hall \(2001\)](#) that uses a log-normal distribution can be used as-is or if it should be expanded to include other types of distributions.

For our initial effort, we used AFM to measure the adhesion force of a 5- μm silica particle (a glass sphere) in contact with two glass slides, one cleaned according to common laboratory standards and another that had been left outdoors for six weeks in the spring of 2015 in Albuquerque, New Mexico. Silica particles and surfaces were chosen for this initial phase because of their sub-nanometer surface roughness and well-defined surface chemistry compared to other particle types (e.g., spores) and urban surfaces (e.g., concrete).

Most of the studies on surface roughness assume a random distribution of asperities over the surface (e.g., [Rabinovich et al., 2000a, 2000b](#); [Prokopovich & Perni, 2010](#); [You & Wan, 2013](#)); however, we will show that the dirty surface is better described in terms of isolated “boulders” in the middle of an otherwise level plain. Since AFM can provide detailed information related to the topography of the surface, the third goal of this paper is to compare the commonly used roughness distributions to our measurements. Finally, the fourth goal will be to estimate the adhesion force distribution from our roughness measurements using the [Prokopovich and Perni \(2010\)](#) model.

The paper is organized as follows: [Section 2](#) describes the experimental set-up; [Section 3](#) presents and discusses the surface roughness and the adhesion force measurements; [Section 4](#) presents the estimation of the adhesion force with a multi-asperity model; and [Section 5](#) contains our conclusions. In the following, we will use the word “clean” to identify the laboratory glass surface, cleaned

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