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airborne dust
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Contribution of airborne dust particles to HONO sources

N. A. Saliba¹, S. G. Moussa², and G. El Tayyar¹

¹American University of Beirut, Department of Chemistry, Beirut, Lebanon

²Air Quality Research Section, Environment Canada, 4905 Dufferin Street, Toronto, Ontario, M3H 5T4, Canada

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Correspondence to: N. A. Saliba (ns30@aub.edu.lb)

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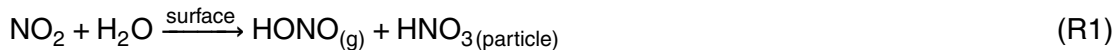
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Abstract

HONO is a major precursor for OH radicals in early mornings. Its formation has been mainly attributed to the heterogeneous hydrolysis of NO₂ on surfaces such as soot, glass, mineral oxides and aerosol surfaces. In particular, dust events which are loaded with mineral oxide aerosols have been associated with higher HONO concentrations in the gas phase. In order to understand the mechanism of reactions related to this process, samples during dusty and non-dusty days were collected between October 2009 and April 2011. Based on HYSPLIT backward trajectories, data were divided between wind trajectories originating from Arabian or African deserts. In this study an increase of HONO levels was observed during dusty days. The increase in the acidic gas concentrations was accompanied by an increase in the PM nitrate and sulfate ion concentrations. During high relative humidity (African dusty days), it is proposed that the mechanism of NO₂ hydrolysis predominates whereas during Arabian dusty days, where the air is relatively dry, a synergistic mechanism of adsorption and reaction between NO₂ and SO₂ on dust particles to produce HONO and sulfate in the particle phase is suggested. This study implies that the NO_x reactivity on mineral oxide surfaces leads to a higher mixing level of OH. An increase in the sulfate forming capacity could account for the underestimation of sulfates in aerosols when the reactive uptake of SO₂ alone is considered.

1 Introduction

HONO is an important precursor for OH radicals in the atmosphere. Compared to ozone and formaldehyde, HONO photolysis is considered the major source of OH radicals in the early morning (Finlayson-Pitts and Pitts, 2000). The main source of HONO formation in the troposphere is attributed to the heterogeneous hydrolysis of NO₂ on wet surfaces (Reaction R1) (Finlayson-Pitts and Pitts, 2000, and references therein).



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This heterogeneous reaction has been studied on several underlying substrates such as soot, glass, mineral oxides and aerosol surfaces (Finlayson-Pitts et al., 2003; Ma et al., 2013; Ullerstam et al., 2003; Underwood et al., 1999, 2001; Yu et al., 2009). In urban regions, the reaction of NO_2 on surfaces is well known to be dependent on relative humidity (Finlayson-Pitts et al., 2003) (and references therein) while the reactivity of the surface towards NO_2 is a function of other competing reactions such as the reaction of NO_2 in the presence of SO_2 on mineral surfaces (Liu et al., 2012). As an example, TiO_2 nanoparticles which have been used in self-cleaning window glass, building materials, and on roads in Europe, Japan and the USA (Beaumont et al., 2009; Langridge et al., 2009) are considered a source for HONO and H_2O_2 from NO_2 and H_2O . TiO_2 in dust was also shown to be a contributor to the formation of HONO (Ma et al., 2013; Ndour et al., 2008). Furthermore, modeling studies suggest that mineral aerosols surfaces account for 40 % of nitrate formation and dust events create a favorable medium for the accumulation of nitrates (Dentener et al., 1996; Guo et al., 2011; Usher et al., 2003). The uptake of SO_2 on mineral oxide surfaces was shown to undergo a two-step mechanism including the reversible adsorption of SO_2 on the surface followed by the oxidation to sulfate (Ullerstam et al., 2002). In the case where SO_2 is co-adsorbed with NO_2 on the surface, it is shown that the oxidant is gaseous NO_2 which reacts with adsorbed SO_2 to produce sulfate in the particle phase. The oxidant role of NO_2 leads to nitrate or contributes to the formation of sulfate on the surface (Borerisen et al., 2000; Ma et al., 2013; Phillips, 2013; Wu et al., 2013).

Given the reactivity of mineral oxide surfaces towards NO_2 and SO_2 , some studies reported an increase of HONO levels during dust storms (Phillips, 2013; Wang et al., 2003; Wu et al., 2013; Zhu et al., 2010), however, the mechanism of HONO formation on airborne dust particles is still not understood. This study assesses the increase of HONO in the gas phase during two dust events; Arabian and Saharan, accompanied with aerosols at different relative humidity. Consequently, two mechanisms of actions for the formation of HONO are proposed.

2 Methods

2.1 Collection techniques

Soluble gases and size resolved aerosols were sampled at ambient level during Arabian and African dusty days. Dusty days were predicted using the NOAA-Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Draxler and Rolph, 2014). The aerosol sampling duration extended over 24 h using Teflon filters housed in Harvard cartridges. Following collection and gravimetric calculation of PM concentrations, the filters are cut into four equal pieces that are frozen until ready for chemical analysis (Kouyoumdjian and Saliba, 2006; Saliba et al., 2009, 2010).

Soluble gases (HONO, HNO₃ and H₂SO₄) were sampled using a diffusion denuder system. The system is based on a series of Honey Comb denuders (HC) for sampling trace gases as described in Saliba and Chamseddine (Saliba and Chamseddine, 2012). The aerosol and gas samples are collected using a Rupprecht & Patashnick Partisol 2300 speciation sampler equipped with ChemComb cartridges in which three Honeycomb (HC) denuders are placed upstream of a pre-weighed Teflon filter that is housed between two Teflon meshes.

2.2 Ion chromatography

Two quarters of each Teflon filter are extracted in 10 mL deionized water and then sonicated for 20 min. Coated denuders are extracted by a 10 mL of a 0.05% H₂O₂ solution. Both extracts are filtered and analyzed by ion chromatography (Saliba and Chamseddine, 2012).

3 Results and discussion

Samples were collected between October 2009 and April 2011. Sampling data were divided into non-dusty and dusty days (Tables S1 and S2). Based on HYSPLIT backward

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trajectories, two routes for wind were identified: a SE backward wind trajectory originating from Arabian deserts and a SW backward trajectory originating from African deserts.

Gas phase composition: The average gas phase HCl, HONO, HNO₃, and H₂SO₄ (sum of SO₂ and H₂SO₄ captured by the denuders) levels for the sampling periods are shown in Fig. 1. The average concentrations for HCl, HONO, HNO₃ and H₂SO₄ during non-dusty days were 1.79 ± 1.11 , 0.89 ± 0.33 , 1.21 ± 0.69 , and $15.69 \pm 5.74 \mu\text{g m}^{-3}$, respectively. There was a burst of HCl, HONO, HNO₃ and H₂SO₄ during dusty days. In the present set of data, a twofold and a threefold increase of HONO levels was observed during African and Arabian dust storms, respectively. The increase in the acidic gas concentration was accompanied by an increase in the PM ions concentration (Fig. 2).

During African dust storms, the relative humidity was high (average RH ~ 75 %). Water molecules form multilayer water films on urban surfaces including particles (Moussa et al., 2009; Sumner et al., 2004). The presence of water on the surface acts as a shield preventing the interaction of NO₂ and SO₂ directly with the active sites on the particle surface. This is confirmed by the correlation between gaseous HONO and HNO₃ ($R = 0.52$) which is enhanced in the Fall and Winter seasons to become 0.94 (Fig. S1). Also, the correlation between NO₃⁻ (particle) and HONO (gas) ($R = 1.00$) during the Fall–Winter season suggests that the heterogeneous hydrolysis of NO₂ on dust particles is the main mechanism for gas phase HONO formation during those episodes. SO₂ reacts with surface water through several steps to form H₂SO₄ which is taken up by the particle phase as SO₄²⁻. This conclusion is confirmed by the positive correlation (0.92) between gas phase HONO and particle sulfate. The noticed higher concentration for HONO in Spring–Summer ($2.2 \mu\text{g m}^{-3}$) relative to the Fall-winter season ($1.9 \mu\text{g m}^{-3}$) suggests an additional photochemical source for HONO due to secondary organic aerosols formation during summer (Broske et al., 2003; George et al., 2005).

The mechanism of NO₂ hydrolysis via the dimerization of NO₂ into N₂O₄ (Reactions R2 and R3) and the formation of NO⁺NO₃⁻ intermediate (Reaction R4) as sug-

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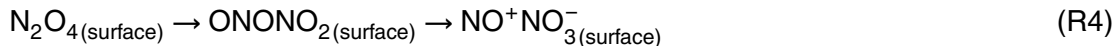
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gested by Finlayson-Pitts and coworkers on water surfaces is proposed (Finlayson-Pitts et al., 2003). The final products are thus HONO_(g) and HNO₃(particle) as illustrated in Reaction (R5).



On the contrary, during Arabian dust storms, where the air is relatively dry (average RH ~ 30%), there was a poor correlation for HONO_(g) with HNO₃(g) ($R = 0.05$) and with HONO_(g) and particulate nitrate ($R = 0.01$) suggesting that Reaction (R1) is not the main source for HONO formation. On the other hand, gas phase HONO was highly correlated with gas phase H₂SO₄ ($R = 0.74$) (Fig. S2), additionally, during the Fall–Winter seasons, there is a strong correlation between particle phase nitrates and sulfates ($R = 0.96$) and during the Spring–Summer seasons, sulfate is highly correlated with H₂SO₄ ($R = 0.92$), HNO₃(g) ($R = 0.67$) and HONO_(g) ($R = 0.95$). These correlations suggest that there is a synergistic mechanism of adsorption and reaction between NO₂ and SO₂ on dust particles; mainly mineral oxides.

Hydroxyl M-OH groups on oxide surfaces react with acidic molecules such as NO₂ and SO₂ via a nucleophilic attack of the oxygen atom to form adsorbed sulfite (M-SO₃) and nitrate (M-NO₃), respectively (Ma et al., 2008; Tsyganenko et al., 2000). When both NO₂ and SO₂ are available, SO₂ competes with NO₂ for the same adsorption sites, and adsorbs more strongly (Ling et al., 2013; Liu et al., 2012; Prades et al., 2009, 2007; Rodriguez et al., 2000). Hence, only a minor amount of NO₂ chemisorbs on the surface to form M-ONO₂ which can proceed to form adsorbed NO₃⁻ via Reaction (R6). Once the surface is pre-saturated with chemisorbed SO₂ (Reaction R7), the majority of NO₂ dimerizes into N₂O₄ which form a physisorbed layer on the surface. The co-adsorption of SO₂ and the oxidant N₂O₄ lead to the formation of H₂SO₄ and HONO in

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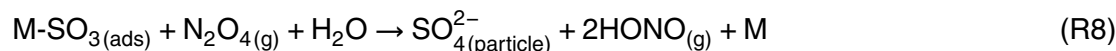
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the gas phase (Reaction R8). Such effect has been shown to exist. For instance, Ma et al. (2008) (Ma et al., 2008) showed that adsorption of NO_2 on alumina was altered in the presence of SO_2 . In similar studies, co-adsorption of SO_2 and NO_2 showed that the intermediate N_2O_4 leads to nitrates and sulfates in pathways different than the formation of NO^+NO_3^- on the surface (Liu et al., 2012). In this proposed model, a competition for NO_2 , between surface adsorption forming nitrate, and its acting as an oxidant, forming sulfate is suggested. Products conform with field measurements of poor correlation between gas phase HONO and gas and particulate phases of nitrates and high correlation between gas phase HONO and gas and particle phase of sulfate. Furthermore, the higher concentration of sulfates in particles during Arabian desert episodes lead us to suggest that the presence of SO_2 on the surface reduces the formation of surface nitrate species while enhancing the formation of surface sulfate.



The enhanced formation of surface sulfate is an important characteristic since it changes the physical properties of the mineral dust particle. A particle coated with sulfate is hygroscopic and will take up water; it may therefore take up more SO_2 into this aqueous layer that would otherwise be formed (Zhang and Chan, 2002). Hence, it is likely that the mineral dust particles have a greater capacity for sulfate formation than suggested by the reactive uptake studies alone (Ullerstam et al., 2003).

The missing source of HONO has several implications on the OH concentration and oxidative budget in the atmosphere. Furthermore, the simultaneous enhanced formation of sulfates in particles impacts their optical and physical properties. Uptake coefficients and rate constants of the proposed reactions can be further developed in laboratory studies so that proposed reactions are incorporated into regional and global models.

Supplementary material related to this article is available online at
[http://www.atmos-chem-phys-discuss.net/14/4827/2014/
acpd-14-4827-2014-supplement.pdf](http://www.atmos-chem-phys-discuss.net/14/4827/2014/acpd-14-4827-2014-supplement.pdf).

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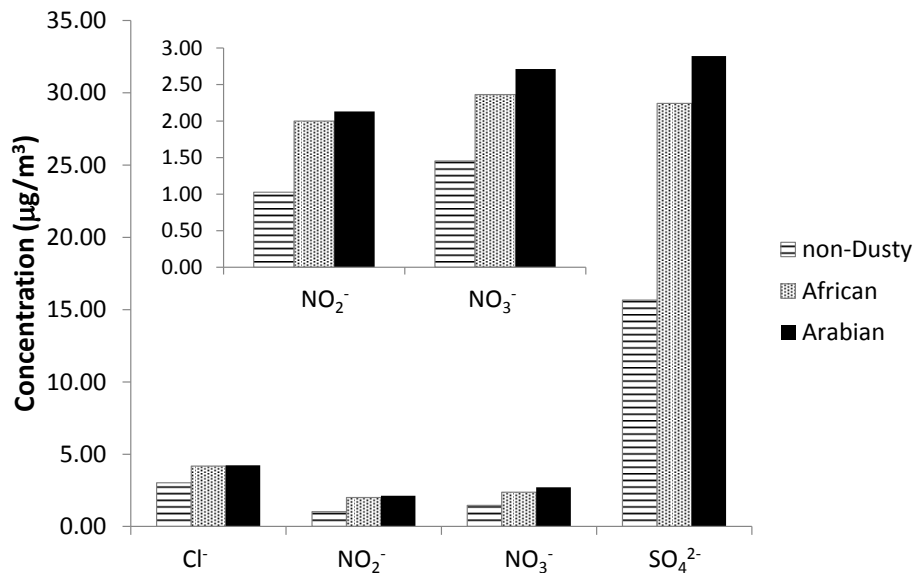


Fig. 1. Average concentration of ions corresponding to gases measured during the sampling period in the presence and absence of dust episodes. The inset expands the y-axis around nitric acid and HONO concentrations.

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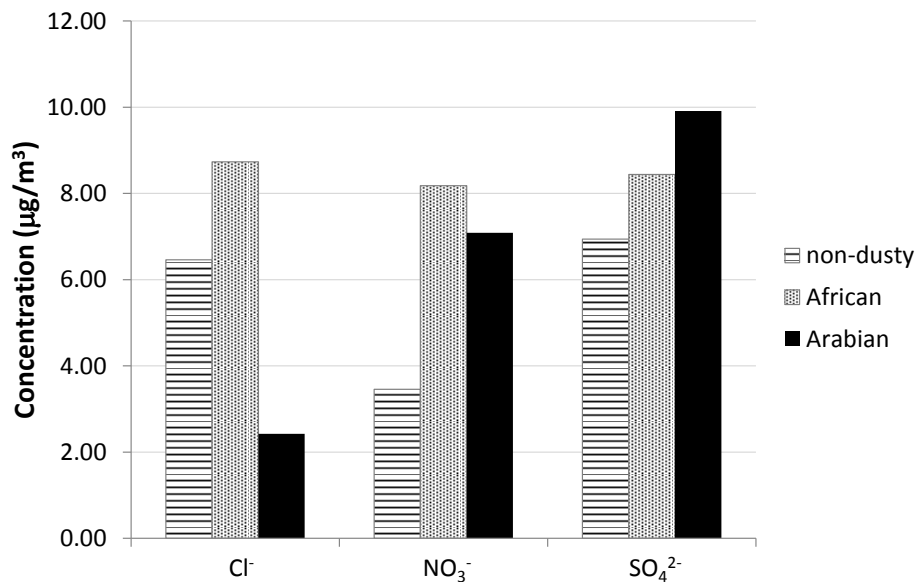


Fig. 2. Average concentration of particle phase chloride, nitrate and sulfate measured during the sampling period in the presence and absence of dust episodes.

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