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The possible role of the surface active substances (SAS) in the airborne transmission of SARS-CoV-2

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\textbf{A R T I C L E A B S T R A C T}

Surface active substances (SAS) have the potential to form films at different interfaces, consequently influencing the interfacial properties of atmospheric particulate matter (PM). They can be derived from both human activities and natural processes and can be found in an indoor and outdoor environment. This paper’s fundamental question is the possible role of the SAS in stabilizing respiratory aerosols in the closed space. In that context, we discuss results of preliminary measurements of the SAS and dissolved organic carbon (DOC) concentrations in the water-soluble fractions of PM\textsubscript{2.5} and PM\textsubscript{10} that were sampled simultaneously in primary school inside and outside of the building. The concentrations of SAS were determined using highly sensitive electrochemical measurements. It was observed that SAS and DOC concentrations have been enhanced indoor in both PM fractions. Consistent with these results, a discussion arises on the possibility that SAS could play a crucial role in respiratory droplet dispersion as stabilizers, especially in a closed space. At the same time, we assume that they could prolong the lifetime of respiratory aerosols and as well viability of some (possible SARS-CoV-2) virus inside of the droplets.

1. Introduction

The review of the literature shows that there is still unknown on the conditions that facilitate the airborne transmission of the new virus-SARS-CoV-2 (Alsved et al., 2020; Asbach et al., 2020; Brlek et al., 2020; Jayaweera et al., 2020; Li et al., 2020; Morawska et al., 2020; Morgenstern, 2020; Tang et al., 2020; WHO, 2020). However, particulate matter especially fine particles (PM\textsubscript{2.5}) have been discussed as a potential SARS-CoV-2 carrier (Jayaweera et al., 2020; Morgenstern, 2020; Nor et al., 2020 and references therein). In the study by Nor et al. (2020) SARS-CoV-2 RNA was confirmed within fine (PM\textsubscript{2.5}) indoor ambient particles sampled in hospital wards with different infected clusters.

The term “airborne transmission” assumes the infections spreading through exposure to the fine solid particles or deliquescent particles containing an infectious virus, at greater distances or over longer times. These particles are originated from respiratory droplets produced by an infectious; after water is evaporated, solid particles are formed by condensation of low volatile compounds (Morawska, 2006; Lindsley et al., 2010, 2013bib_Lindsley_et_al_2010). A previous study has reported that the oral cavity is the major source of expiratory droplets of which the vast majority during speech and coughing is less than 2 \(\mu\text{m}\) (Johnson and Morawska, 2009).

Atmospheric particles in the Aitken (smaller than 100 \(\text{nm}\)) and accumulation mode (100–1000 \(\text{nm}\)) (Seinfeld and Pandis, 2006) have a shorter relaxation time and stopping distance, and consequently can remain suspended for several days, resulting in a long-range transport (Willeke and Whitby, 1975; Hinds, 1999; Moris et al., 2015; Leonardi et al., 2020 and references therein). On the other hand, aerosols, as well as other colloidal dispersions (particles ranging from 1 \(\text{nm}\) to 1 \(\mu\text{m}\)), are thermodynamically unstable systems because they tend to minimize surface energy by coagulation of particles and finally by their sedimentation (Goodarzi and Zendehboudi, 2019, and references therein).

The heterogeneous systems with phase boundaries could be stabilized by surface active substances (SAS) defined by amphiphilic nature (hydrophobic and hydrophilic segments). Therefore, they tend to adsorb at the phase boundaries such as liquid/liquid, liquid/solid, liquid/gas, or solid/gas (Attwood and Florence, 1983; Myers, 1999). By adsorption and solubilization, SAS may influence physico-chemical properties and structure of natural interfaces, and in that way moderate transfer processes between different phases. Depending on the adsorbate and nature

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https://doi.org/10.1016/j.envres.2021.111215
Received 10 December 2020; Received in revised form 18 March 2021; Accepted 20 April 2021
Available online 30 April 2021
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of the interface, adsorption of SAS can be influenced by hydrophilic, electrostatic hydration, and hydrophobic interactions (Conway, 1976; Westall, 1987). Surface active compounds are found in a different part of the environment as a product of natural processes and human activities (Olkowska et al., 2014; Renard et al., 2016; Orlovic-Leko et al., 2016; Ciglenecki et al., 2020). The latter is very often, especially in the case of detergents, less abundant in mass while on the contrary, they may represent the most surface-active organic material in the different environmental compartments (Olkowska et al., 2014).

SAS were confirmed in various atmospheric samples (cloud water, rainwater, snow, aerosol particles) (Facchini et al., 1999; Orlovic-Leko et al., 2004, 2009, 2010, 2020; Renard et al., 2016; Krollic et al., 2018; Gérard et al., 2019; Cvitešić Kusan et al., 2019). In microscopic systems with relatively large surface areas, such as atmospheric droplets and deliquescent particles, adsorption of SAS at interface solutions/air is highly important (Morris et al., 2015; Malila and Prisle, 2018; Lin et al., 2020; Bzek et al., 2020). The presence of SAS could cause significantly reducing the surface tension of droplets. That is relevant for increasing the population of droplets of smaller size which are more easily transported from one place to another (Bzek et al., 2020). Thereby, the surface tension of aqueous particles is discussed as a function of the relative humidity (RH) in the atmosphere (Morris et al., 2015; Bzek et al., 2020); smaller particles need a higher ambient RH to maintain equilibrium than larger ones.

The SAS have been detected in the indoor air as well (Morawska et al., 2003, 2006; Wolkoff et al., 1998; Ahmad et al., 2009). The surfactants derived from cleaning agents can irritate the human respiratory system leading to allergies and asthma as well as to dried eyes by reducing the surface tension of the tear film (Olkowska et al., 2014 and references therein). Once inhaled, atmospheric surfactants may interact with a pulmonary surfactant, i.e., those that cover lung alveolar surface and include polar phospholipids and hydrophobic specific low-molecular proteins (Exerowa et al., 2014). The pulmonary SAS, which is crucial for the normal breathing function, could also contribute to the dissolution of aerosol particles (Brimblecombe and Sukhapan, 2002).

As stated, respiratory aerosols are created when air passes over a layer of fluid (Fiegel, 2006; Morawska, 2006, 2009). The surfactant increase in overall droplet formation has been already discussed as a cause for smaller droplets. The small particles (Aitken and accumulation mode, 10–1000 nm) least likely impact and settle on surfaces and can float on the air and spread much further following the airflow stream especially after being dried (Lindsey et al., 2013).

Consistent with these considerations, in this paper, we discuss the possibility that the organic SAS could play a significant role in stabilizing the “cloud of these respiratory particles”. We assume that SAS’s presence could prolong bioaerosols’ lifetime, protecting particles from the deposition process. At the same time, SAS that are adsorbed on the respiratory droplets bearing virus can keep it for a longer time viable. According to the above discussion, it could be expected that SAS can act in respiratory droplets dispersion as a stabilizer, which can result in their persistence in the air for more extended periods, especially in the closed environment. As support to our assumption, we present results of total SAS and DOC concentrations in the water-soluble organic carbon (WSOC) fraction of PM$_{2.5}$ and PM$_{10}$ collected parallel in the primary school’s outdoor and indoor environment. These measurements confirmed up to two times higher concentrations of the SAS in the indoor samples.

2. Methodology

2.1. Sampling and preparing analyte solution

The PM$_{2.5}$ and PM$_{10}$ were sampled on the Quartz fiber filters (Whatman QMA, 47 mm diameter) at the primary school located in the city center, from 8 a.m. to 8 p.m. (teaching hours) and from 8 p.m. to 8 a.m. (no teaching hours) (Kovacev et al., 2015). The indoor and outdoor measurements were carried out at the same time using low volume (2.3 m$^3$/h) samplers, LVS3 (Sven/Leckel LVS3) with size-selective inlets for PM$_{10}$ and PM$_{2.5}$ fractions. PM concentrations were obtained from gravimetric analysis of sampled filters. Indoor samples were taken in the hallway with a floor space of 60 m$^2$ and a volume of 200 m$^3$. The hallway was occupied with an average of 80 pupils during the breaks between classes, while ambient air was sampled on the balcony about 10 m above the ground. There was no additional ventilation system in the building. Pre-conditioning and post-conditioning of filters were undertaken in accordance with the general requirements of EN 12341:2014. Approximately 15% of all gravimetric samples were collected as field blanks. After preconditioning in a clean room, filters were weighing using the Mettler Toledo semi-micro balance (with min. 10 μg mass resolution). PM concentrations were calculated using the average (each filter is measured three times) weight of filters.

The water-soluble organic components (WSOC) of atmospheric indoor particles were extracted by placing half of the filters in 25 ml of MilliQ water (Millipore Corp.) for 24 h and filtered through 0.7 μm GF/F filters (Whatman, 47 mm diameter). In this analyte solution, the SAS was quantified by highly sensitive electrochemical measurements (EM) of the adsorption effect at the mercury electrode.

2.2. Electrochemical measurements

The study was performed by highly sensitive electrochemical in-house methodology (Cosovic and Vojvodic, 1989) developed for determination of SAS concentration in different water samples (Orlovic-Leko et al., 2004, 2004, 2009, 2010, 2016, 2020; Cosovic et al., 2007; Ciglenecki et al., 2020 and references therein) by the phase sensitive alternating current (AC) voltammetry, PSAVC (out-of-phase signal, frequency 77 Hz, amplitude 10 mV). Electrochemical analyzer µAutolab-type (Eco Chemie B.V., The Netherlands) equipped with GPES 4.6 software (Eco Chemie B.V., The Netherlands) was used. Adsorption effect of SAS was measured at the hanging mercury drop electrode (HMDE, Metrohm, Switzerland) of the surface area A = 0.022 cm$^2$. The measured potentials are reported with respect to the Ag/AgCl (3 M KCl) electrode. The base electrolyte was 0.55 M NaCl. The concentration of SAS was expressed as equivalent (eqv.) in mg L$^{-1}$ to the nonionic surfactant, polyoxyethylene-t-octylphenol (Triton-X-100, Rohm and Hass, Milano, Italy) based on external calibration by using Triton-X-100 calibration curve (conc. range between 0.01 and 1 mg L$^{-1}$) in 0.55 M NaCl (Cosovic et al., 2007; Orlovic-Leko et al., 2020). Triton-X 100 is likely to represent many atmospheric SAS, as its critical micelle concentration (CMC and γCMC) is consistent with those reported in atmospheric PM (Leonardi et al., 2020). The limit of detection (LOD) for voltammetric SAS determination was 0.01 mg L$^{-1}$ eqv. of T-X-100, with limit of quantification (LOQ) of 0.03 mg L$^{-1}$.

In addition, WSOC content, i.e. DOC was determined by the high-temperature catalytic oxidation (HTCO) method at a TOC-VCPH instrument (Shimadzu, Japan) (Cosovic et al., 2007; Cvitešić Kusan et al., 2019). The WSOC sample aliquot (15 mL) was acidified with 2 M HCl to pH ~ 3 in order to eliminate the inorganic carbonates. The concentration of the adsorption effect at the mercury electrode. The base electrolyte was 0.55 M NaCl. The concentration of SAS was expressed as equivalent (eqv.) in mg L$^{-1}$ to the nonionic surfactant, polyoxyethylene-t-octylphenol (Triton-X-100, Rohm and Hass, Milano, Italy) based on external calibration by using Triton-X-100 calibration curve (conc. range between 0.01 and 1 mg L$^{-1}$) in 0.55 M NaCl (Cosovic et al., 2007; Orlovic-Leko et al., 2020). Triton-X 100 is likely to represent many atmospheric SAS, as its critical micelle concentration (CMC and γCMC) is consistent with those reported in atmospheric PM (Leonardi et al., 2020). The limit of detection (LOD) for voltammetric SAS determination was 0.01 mg L$^{-1}$ eqv. of T-X-100, with limit of quantification (LOQ) of 0.03 mg L$^{-1}$.

3. Results and discussion

Results of the voltammetric measurement of SAS in the WSOC fraction of atmospheric PM$_{2.5}$ and PM$_{10}$ samples collected simultaneously in the indoor and outdoor air of the school environments were presented in Fig. 1 and Table 1. Additionally, in Table 1, the DOC concentrations and relevant data about PM samples were listed (Kovacev et al., 2015).

As can be seen from Table 1, indoor concentrations of PM$_{2.5}$ collected during school teaching hours were higher (26.6 μg m$^{-3}$) than outdoor
concentrations (20.0 μg m⁻³) while the PM₁₀ concentrations found in classrooms, during no teaching hours, were slightly lower (40.0 μg m⁻³) than in outdoor air (47.4 μg m⁻³). Kovacević et al. (2015) concluded that the high outdoor PM concentrations and resuspension of particles could be the main possible reasons for the elevated indoor PM concentrations.

Generally, in a closed space without strong particle sources, the indoor PM would be expected to be the same as, or lower than, outdoor levels. In addition, indoor PM levels have the potential to exceed PM levels in the ambient air. This observation could be coupled to the particles' generation by specific sources and/or personal activities of occupants. However, indoor PM can also be of biological origin (Cox et al., 2020).

In this work, the analyte solutions prepared from the same aerosol samples (Table 1), quantification of SAS was done by using calibration curve of the Triton-X-100 (external calibration) (Cosovic et al., 2007) and measured values of ΔI [nA] (Fig. 1) from the recorded AC voltamograms. The decrease in the capacitive current value in the analyte samples, concerning the current of the supporting electrolyte (ΔI), is a direct measure of the surface-active organic material adsorbed at the working (in our case Hg) electrode. It is evident that the surface activity of the WSOC fraction of PM indoor (PM₂.₅ = 0.24 and PM₁₀ = 0.25 mg L⁻¹ eqv. Triton-X-100) was up to two times higher than that of the PM outdoor samples (PM₂.₅ = 0.15 and PM₁₀ = 0.12 mg L⁻¹ eqv. Triton-X-100). Accordingly, higher DOC concentrations were measured in PM WSOC fractions of the indoor samples (Table 1), indicating that the indoor air contained 40% more DOC. However, in this preliminary set of measurements, the difference in the indoor SAS concentrations during teaching and not teaching hours was not expressed. Considering that the school is located in a street with heavy traffic, it could be expected that indoor SAS level would be affected by traffic emissions, i.e., by compounds that migrate from the ambient air. However, our results point to the importance of the indoor generated SAS fraction of DOC. The cleaning agents with surface-active components are likely to be crucial in explaining this observation, especially after teaching hours, when cleaning of classrooms is usually performed. It is known that household cleaning activities and evaporation processes of semi-volatile compounds (anionic and nonionic surfactants) from different surfaces can be a significant source of SAS in the closed environment (Ahmad et al., 2009; Olkowski et al., 2014). In addition, several other studies suggest that in the indoor environment, the SAS derived from cleaning agents could be argued as agents promoting sick-building syndrome (Wolkoff et al., 1998; Sukhapan and Brimblecombe, 2002). Disinfectants are especially highlighted as the most dangerous group of cleaning agents which significantly contribute to the SAS pool (Wolkoff et al., 1998).

This study's general idea is that indoor generated SAS could stabilize bio-aerosols and influence its microphysics processes. Due to their dual nature, SAS with molecules that contain hydrophobic and hydrophilic parts tend to adsorb at different phase boundaries. Adsorption of SAS at the solutions/air interface is significant for small atmospheric droplets due to their relatively large surface areas (Prisle et al., 2010; Malila and Prisle, 2018; Lin et al., 2020; Bzdek et al., 2020). The lower the droplet's radius or, the higher the surface to volume ratio, the higher the WSOCC concentration in organic PM was measured (Ervens and Volkamer, 2018). The specific properties of SAS could cause a reduction of surface tension of droplets (an essential factor for their vapor pressure) and, in that way, increase and stabilize the population of droplets of the smaller sizes. Such SAS coated droplets or deliquescent particle could remain suspended longer in the air (Tessum et al., 2014; Bzdek et al., 2020). Additionally, it is essential to know that SAS can either enhance or slow down the transfer of water across the surface according to the hydrophilic or hydrophobic nature of the aerosol organic coating. These organic coats are common on aerosol particles and might retard the evaporation of molecules present in the water phase, reduce gas transfer, influence chemical reactions, and alter the absorption or reflection properties of aerosols (Remard et al., 2016; Bzdek et al., 2020). This kind of thinking certainly goes in favor of observations that the SARS-CoV-2 virus has spread much faster in the indoor environment.

4. Conclusion

The preliminary results of highly sensitive AC voltammetry out-of-phase measurements of the indoor and outdoor SAS concentrations in the WSOC fraction of the atmospheric PM₂.₅ and PM₁₀ sampled inside and outside of the primary school building, during and after school time, have been shown that indoor SAS concentration was almost two times higher compared with the outdoor.
higher than those found in the outdoor air. Such results indicate that indoor SAS could have different sources than outdoor, as already reported (Ahmad et al., 2009). We have assumed that SAS derived from cleaning agents can be the main reason for the observed higher indoor SAS levels, especially after teaching hours. However, biosurfactants from bioaerosols (Cox et al., 2020) that are expected to be enhanced during teaching hours by breathing, speaking, singing, coughing, sneezing when children are in school, could also be a critical source of dissolved organic carbon with surface active properties (SAS). These surface-active organic carbon molecules, present in a sufficient concentration, can potentially act as a stabilizer of the cloud of respiratory droplets in the indoor air/environments. SAS that are adsorbed on the respiratory droplets bearing virus (potentially SARS-CoV-2) can prolong their lifetime in the indoor air, and in that way may significantly contribute to airborne transmission of COVID-19. Further research to prove posted assumptions in this paper is planned, including analyses of fine (PM$_{2.5}$ and smaller) indoor aerosols from different community locations (schools, gym, first aid stations) on possible SARS-CoV-2 presence.

Author contribution

Irena Ciglenečki and Palma Orlović-Leko: conceptualization, writing – original draft, writing – review & editing. Irena Ciglenečki: idea, resources, funding acquisition. Kristijan Vidović: discussion, writing – review & editing. Vasa Tasić: sampling, manuscript editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work arose from discussions on organic matter characterization in natural samples including marine aerosols, during the implementation of the Croatian Science Foundation project: IP-2018-01-1717, MARRES. The authors thank A. Cvitvić-Kusian for part of the SAS measurements; T. Lovrinović for help in designing of graphical abstract, and the anonymous reviewers for their valuable comments that improved the manuscript.

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