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Cite this article: Fedorets AA, Bormashenko E, Dombrovsky LA, Nosonovsky M. 2019 Droplet clusters: nature-inspired biological reactors and aerosols. *Phil. Trans. R. Soc. A* **377**: 20190121.
http://dx.doi.org/10.1098/rsta.2019.0121

Received: 7 March 2019
Accepted: 9 April 2019

One contribution of 16 to a theme issue 'Bioinspired materials and surfaces for green science and technology (part 2)'.

Subject Areas:
fluid mechanics, chemical physics, materials science

Keywords:
droplet cluster, breath figure, bioaerosol, self-assembly

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THE ROYAL SOCIETY PUBLISHING

Droplet clusters: nature-inspired biological reactors and aerosols

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Condensed microdroplets play a prominent role in living nature, participating in various phenomena, from water harvesting by plants and insects to microorganism migration in bioaerosols. Microdroplets may also form regular self-organized patterns, such as the hexagonally ordered breath figures on a solid surface or levitating monolayer droplet clusters over a locally heated water layer. While the breath figures have been studied since the nineteenth century, they have found a recent application in polymer surface micropatterning (e.g. for superhydrophobicity). Droplet clusters were discovered in 2004, and they are the subject of active research. Methods to control and stabilize droplet clusters make them suitable for the *in situ* analysis of bioaerosols. Studying life in bioaerosols is important for understanding microorganism origins and migration; however, direct observation with traditional methods has not been possible. We report preliminary results on direct *in situ* observation of microorganisms in droplet clusters. We also present a newly observed transition between the hexagonally ordered and chain-like states of a droplet cluster.

This article is part of the theme issue 'Bioinspired materials and surfaces for green science and technology (part 2)'.

1. Introduction

Water microdroplets play a significant role in various processes in nature, ranging from fog and cloud formation to water harvesting by plants and insects, as well as natural self-cleaning and slippery surfaces. Microdroplets also form aerosols and bioaerosols, which can transport various microorganisms for extended distances.

Tracking life in bioaerosols is important for understanding the biodiversity and migration of microorganisms; it may also be relevant to problems of the origin and early stages of life on Earth. However, direct *in situ* observation of microorganisms in airborne bioaerosols is almost impossible, because it is very difficult to trace individual microdroplets. Consequently, questions like for how long a microorganism can survive in an isolated airborne microdroplet or whether cell reproduction is possible while flying, can currently be answered only in an indirect manner.

In some cases, microdroplets form ordered arrangements, such as self-assembled breath figures and droplet clusters. The breath figures are patterns formed by droplets on a relatively cold smooth solid surface (for example, on glass) when water vapour contacts such a surface [1]. Although breath figures have been known for a long time, their new applications for the synthesis of micropatterned polymeric surfaces have been discovered relatively recently, which stimulated renewed interest towards this phenomenon in the community of materials scientists and surface scientists working with polymers. Micropatterned polymeric surfaces can be used, for example, for the synthesis of superhydrophobic water-repellent materials using the lotus effect.

The phenomenon of microdroplets levitating over a locally heated layer of water or other liquids in an ascending vapour–air flow and forming a hexagonally ordered monolayer called a 'droplet cluster' has been reported for the first time in 2004 [2]. The droplet cluster became a topic of extensive research. It was demonstrated that the forces responsible for the formation of the cluster are of aerodynamic nature rather than electrostatic [3]. A feasible method to generate a cluster with any desired number of monodisperse droplets has been suggested [4]; also methods to control droplet size and the distance between them, to trace the droplets, and to use them as chemical and biological reactors have been suggested [5].

The droplet cluster is similar to phenomena where self-organization is prominent, such as water breath figures, colloid and dust crystals, foams, Rayleigh–Bénard cells, or even Wigner crystals. While self-assembled droplet clusters have not been observed in nature, they may be related to phenomena such as long-term cloud stability. Droplet clusters also have a connection to biomimetics, since ordered patterns of condensed droplets are observed, for example, during water harvesting in living nature.

In this paper, we review and present recent results on the self-organized breath figures and droplet clusters. We will discuss their relation to other phenomena involving droplet self-assembly, such as water harvesting in nature. We will also present new results about droplet clusters' application for biological research and the transition between the ordered arrangement and a chain arrangement in these clusters.

2. Droplet-wise condensation in nature: breath figures and water harvesting

A well-known example of water condensation in nature is dew forming on cold objects, whose temperature is below the dew point for a given relative humidity of the air. However, in some cases, dew formation is combined with self-organization of droplet patterns. These are the breath

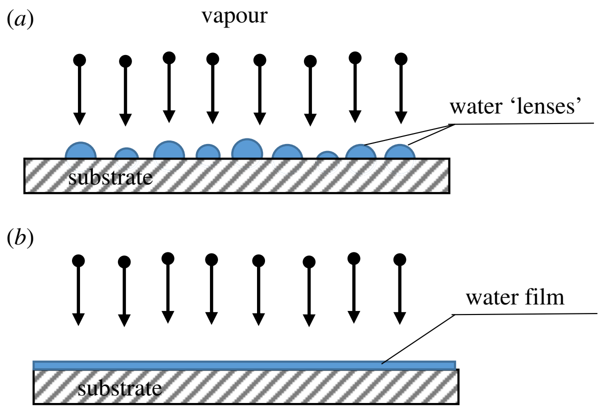

Figure 1. Drop-wise (*a*) and film-wise (*b*) scenarios of condensation depicted schematically. (Online version in colour.)

figures, which can be used for various applications, ranging from photography, to polymer surface patterning, and to water harvesting.

(a) Droplet patterns in self-organized condensed breath figures

The term ‘breath figures’ refers to the fog consisting of microdroplets on a solid surface that forms when water vapour touches a cold surface [1]. This is a commonly observed phenomenon in daily life, for example, fog that appears on a window when one breathes on it, which explains the term ‘breath figures’.

Ancient scholars paid attention to the importance of water and its relation to air. Thus, Thales considered water as the first principle of all things or αρχη (*arche*, ‘first element’ out of which everything is made) [6]. Breath figures condensation thus exemplified the *arche* of nature, combining breath, air and water. In the modern era, a systematic study of the breath figures was carried out by Aitken [7], Rayleigh [8] and Baker [9].

Aitken distinguished two types of water condensation: film-wise and drop-wise. He discovered that film-wise water condensation took place on a glass surface treated with a blow-pipe flame, whereas drop-wise condensation occurred on a non-treated surface of the same glass. Drop-wise and film-wise scenarios of condensation are schematically depicted in figure 1. Examples of these two condensation pathways are shown in figure 2, representing the environmental scanning electron microscope (ESEM) images of condensing water on a pigeon’s feathers and on metallic surfaces. Obviously, only drop-wise condensation gives rise to breath-figure patterns.

The factors determining whether condensation will occur in the drop-wise or the film-wise manner remain obscure. Similarly, it is not completely clear what prevents droplets from coalescence and what controls their size and the equilibrium distance between them. However, it is generally agreed that heat transfer is crucial for this switching, and consequently it is of primary importance for constituting the breath-figure patterns [10]. Several hypotheses have been proposed to explain why water droplets do not coalesce and have the same size and ordered arrangement. The Marangoni convection caused by a temperature gradient along with the evaporating vapour may suppress the coalescence and control the droplets’ size and the distances between them.

Contamination plays a role in the formation of the breath-figure patterns. While Aitken pointed out the role of dust contamination [7], Rayleigh and Baker emphasized the importance of grease for patterning. The grease was removed by the blow-pipe flame in the experiments reported by Aitken, thus influencing the resulting breath-figure patterns [8,9]. Baker also addressed the electrical breath figures already considered by Aitken [7]. Both Aitken and Rayleigh, as well as Baker, indicated that the breath-figure patterns originate from the complicated

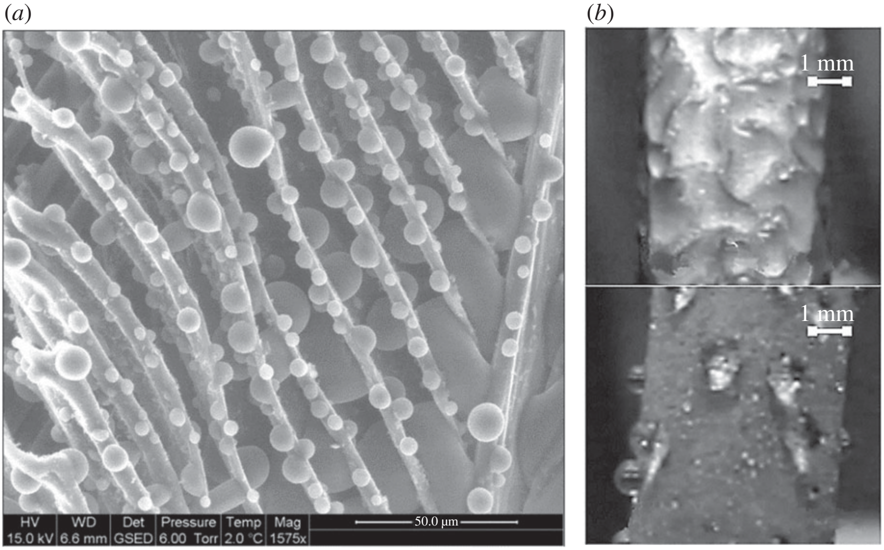

Figure 2. ESEM image of (*a*) the drop-wise condensation of water on a pigeon's feather is depicted. (*b*) Film-wise (the upper image) and drop-wise condensation (the lower image) of water vapour on metallic surfaces.

interplay of the physical (mass and heat transfer) and interfacial effects (presence of grease and solid contaminants on the surface) exerted on the water vapour.

An early application of the breath figures in the nineteenth century was the wet-collodion technique for photographic plates, where glass cleanliness was extremely important. A uniform breath-figure film was formed on a clean surface, while a drop-wise arrangement indicated oil contamination.

Half a century later interest in breath-figure formation was revived in view of the study of atmospheric processes, and in particular the extended study of dew formation, which turned out to be a complicated physical process. The experimental and theoretical study of dew formation has been carried out by Beysens *et al*. [11]. A new application of the breath figures was discovered in 1994, when Widawski, François and Pitois reported manufacturing porous polymer films with a self-organized pattern of micro-scaled honeycomb morphology, synthesized using the breath-figure condensation process [12]. The process involved rapidly evaporated polymer solutions exposed to humidity [1,13–15]. The evaporation of solvent decreased the surface temperature of the solution, thus promoting the drop-wise condensation of water vapour. Water droplets 'drilled' micro-scaled holes, and the residual polymer fixed the honeycomb pattern as depicted schematically in figure 3. A typical breath-figure-induced image is depicted in figure 4*a*. There are numerous applications of porous structures arising from the self-assembly of breath figures, including superhydrophobic surfaces [16].

One of the still unexplained features of breath-figure self-assembly is the hierarchical topography of the eventual pattern with the large scale of 10–50 µm, shown in figure 4*b*, and the small scale of 0.2–2.0 µm illustrated in figure 4*a* [17–22]. Detailed understanding of the breath-figure self-assembly mechanisms remains a challenging task.

(b) Water harvesting by insects and plants

Dew-like water condensation can lead to pattern formation also in various biological processes such as water harvesting. Several species of flora and fauna found in arid areas have developed unique structures for water harvesting from fog and moist air. In most cases, the pattern of liquid is a result of the interaction of vapour with a pattern on a solid surface. One well-known

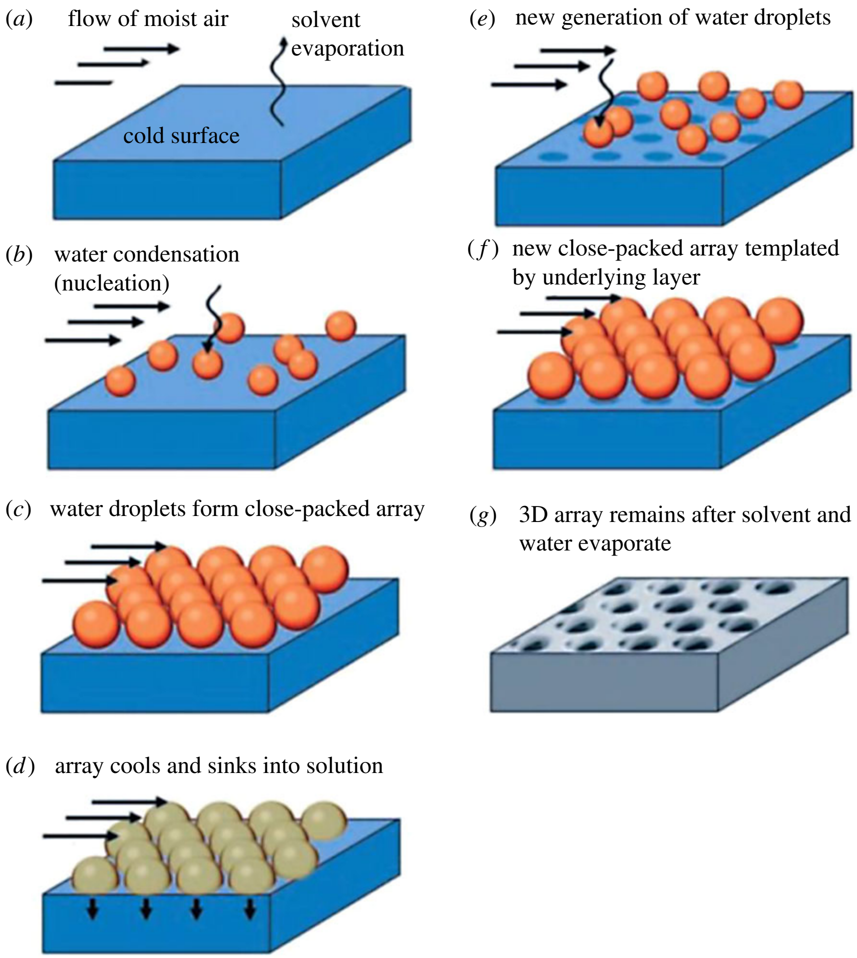

Figure 3. (*a*–*g*) The scheme of a breath-figures process performed with rapidly evaporated polymer solution. (Online version in colour.)

example of water harvesting is supplied by the darkling beetles of the arid Namib Desert in southwestern Africa, one of the driest habitats in the world. Darkling beetles can gather water, which condenses on their body surface from dew and ocean fog [23,24]. Micro-sized relief including bumps and grooves on the beetle's forewings, which includes a combination of hydrophobic and hydrophilic spots, promotes water condensation and directs water towards the beetle's mouth.

Besides insects, desert plants can also be involved in fog and dew harvesting. This includes *Dryopteris marginata* fern [25], *Syntrichia caninervis* moss [26], bushman grass *Stipagrostis sabulicola* [27] and *Opuntia microdasys* cactus [28,29]. In many of these cases, hierarchical organization of water-harvesting patterns plays a crucial role for water collection, including specialized structures in plants [30,31] and structures created by insects such as spider webs [32]. Figure 5 shows dew water droplets and films coexisting by vapour condensation on a spider web.

3. Self-organized levitating droplet clusters

Levitating monolayer clusters of condensed microdroplets is another phenomenon where self-organization of condensed droplets is prominent.

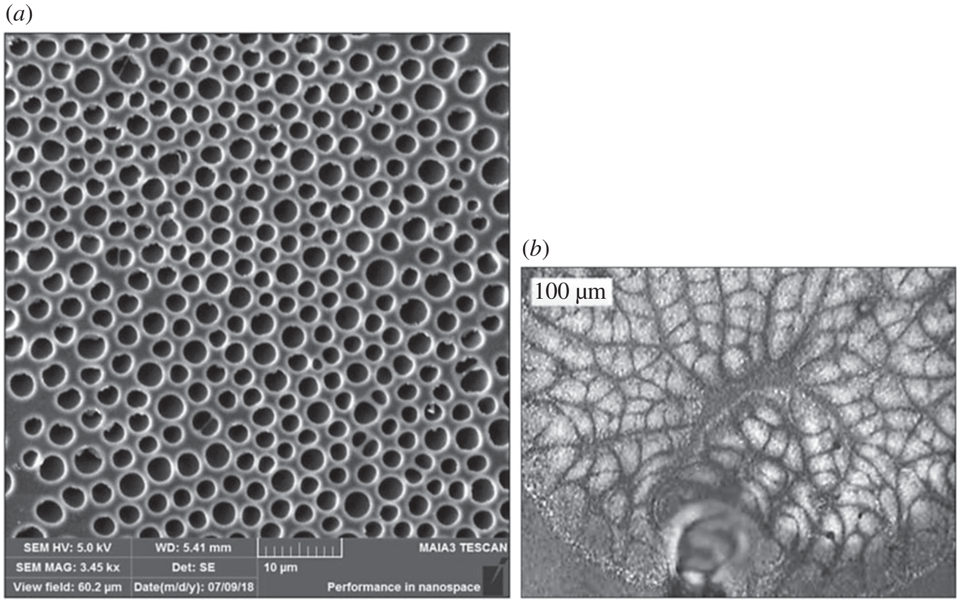

Figure 4. (*a*) SEM image of a typical breath-figures-based pattern obtained with polystyrene dissolved in a mixture of chlorinated solvents. (*b*) The large-scale pattern observed under the breath-figures self-assembly.

Figure 5. Morning mist condensed on spider's web silk. Both stable drop-wise and film-wise structures are observed.

(a) The phenomenon of levitating droplet cluster

To generate a levitating droplet cluster, a water layer with the typical thickness of about 0.5 mm is usually used. A spot on the surface of the layer can be heated by a laser beam or another source of heat to the temperatures of 60–95°C, which results in an air–water flow rising above the heated spot. Owing to the temperature gradient, spherical droplets with almost identical

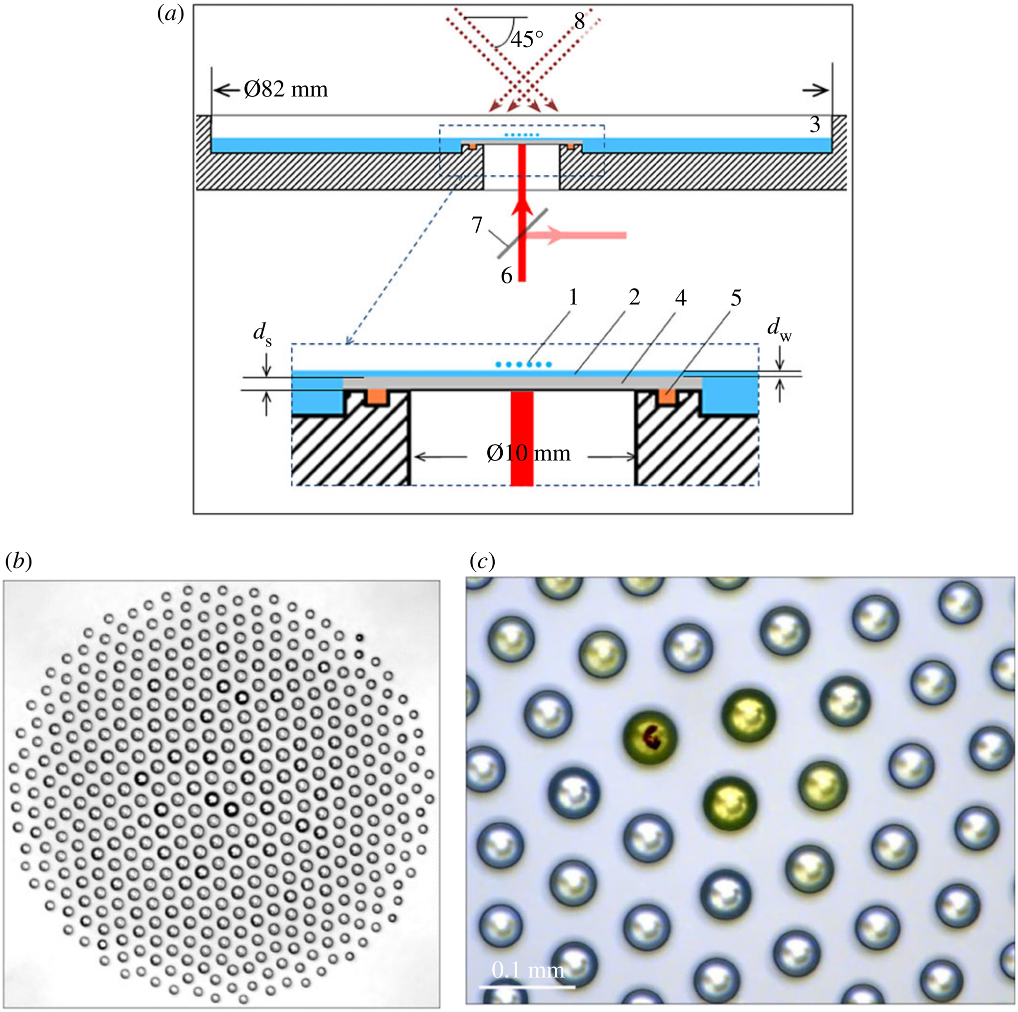

Figure 6. (*a*) The experimental set-up for the droplet cluster. (*b*) A hexagonally ordered droplet cluster. (*c*) A droplet cluster with a non-volatile water-soluble yellow dye injected into several droplets [33]. (Online version in colour.)

diameters (usually between 0.01 mm and 0.05 mm) condense and levitate in the ascending air–vapour flow. Droplets with similar diameters self-organize into a hexagonally ordered monolayer, while smaller droplets tend to form at the periphery of the cluster, where the temperature and the intensity of the rising gas stream are lower than those in the central region (figure 6). The 2D ordering of droplets can be quantified with the Voronoi entropy [4,34]. The phenomenon has also been observed with liquids other than water, including glycerol, benzyl alcohol and ethylene glycol [2–5,34].

The schematic of the experimental configuration is presented in figure 6*a*, where 1 is the droplet cluster, 2 is the horizontal water layer, 3 is the cylindrical cavity in the duralumin plate with the central orifice, 4 is the sitall plate with an absorbing opaque coating, 5 is the annular groove filled with epoxy glue, 6 is the laser beam used to heat the substrate, 7 is the special plate to reflect a part of the laser radiation and 8 are the sources of infrared radiation.

The vertical component of the drag force in the ascending flow is equilibrated by the droplet's weight at a certain equilibrium levitation height. This height is usually of the same order as the droplets' radius. While the droplets are dragged towards the centre of the heated spot, they do not merge with each other due to an aerodynamic repulsive pressure force from the gas flow between the droplets. Note that unlike in the classical Bernoulli problem where the aerodynamic force between two bodies in a flow around them is always attractive, the interaction between two microdroplets is quite complex and can be repulsive [3,34]. The repulsive force is especially important because the diameter of ordinary droplet clusters is much greater than the height of the cluster levitation. This situation is quite different from the wide flow around two side-by-side droplets.

The balance of the repulsion force between the droplets and of the horizontal component of the drag force, which drives the droplets in the direction towards the centre of the heated spot, where the temperature and the intensity of the ascending flow are the highest, leads to the densest packing arrangement of the droplet monolayer (figure 6*b*). This is similar to packing of identical rigid balls at a bottom of a bowl. The densest packing of identical spheres on a plane is provided by the hexagonal (honeycomb) arrangement.

It is possible to inject various substances into droplets, which make them appropriate for biochemical microreactors. Thus, figure 6*c* shows a droplet cluster with a non-volatile water-soluble yellow dye injected into several droplets [33].

For a long time, it remained controversial whether droplets in the cluster can bear an electric charge and whether the electrostatic repulsion between them is significant to prevent their merger and coalescence. However, recent experiments with the cluster in an external electric field of different polarities have shown that the effect of the electrostatic force is negligible in comparison with that of the aerodynamic force [3]. Numerical simulations with the Langevin method taking into account droplet repulsion and random diffusion have also shown results similar to the experimental observations [35].

Normally, condensed droplets would grow with their diameters increasing proportionally to the square root of time [11]. The growth will continue until the droplet touches the water layer and coalesces, which triggers a capillary wave on the water surface, which destroys the entire cluster in a chain reaction. However, it is possible to suppress the growth of droplets by heating them with infrared radiation. This shifts the equilibrium of the evaporation and condensation in such a way that droplets can be stabilized with constant diameters for extended periods of time (hours) [5]. By controlling the temperature and temperature gradient, one can control the number of droplets and their density and size as well as generate a cluster with a desired number of droplets.

While temperatures close to the water boiling point are not appropriate for experiments with living cells, the droplet cluster can also be observed at much lower temperatures of about 20°C [36]. Note that the phenomenon of the droplet cluster is different from the Leidenfrost effect because the latter occurs at much higher temperatures over a solid surface, while the droplet cluster forms at lower temperatures over a liquid surface. While the cluster can be observed even over a dry solid island surrounded by water, the mechanism of levitation is likely to remain dominated by aerodynamic drag of the ascending flow.

Another recent development is the ability to generate small monodisperse clusters consisting of any desirable number of droplets. To generate clusters with a constant number of almost identical droplets (i.e. with very close diameters), the process should be divided into two stages. At the initial assembly stage, the power output of the heating laser was maintained at the small level, which allowed the droplets to migrate towards the centre of the heated spot. After the required number of droplets was obtained, the power output was abruptly increased, leading to the increased speed of the ascending vapour–air flow. The droplets generated in the heating zone at the initial stage were large enough to levitate, while newly created small droplets were blown away by the strong vapour–air flow. Consequently, the cluster contained only those droplets which were generated at the initial stage. Since the cluster assembly stage was short, the diameter of the droplets was uniform [4].

A modified experimental set-up can be used to generate the droplet cluster at room temperatures, which is desirable for the study of biochemical processes in the droplets. Such a modified configuration includes a separate volume of cold air flow above the central part of the water layer. Even moderate local heating of the water surface is the most important factor to

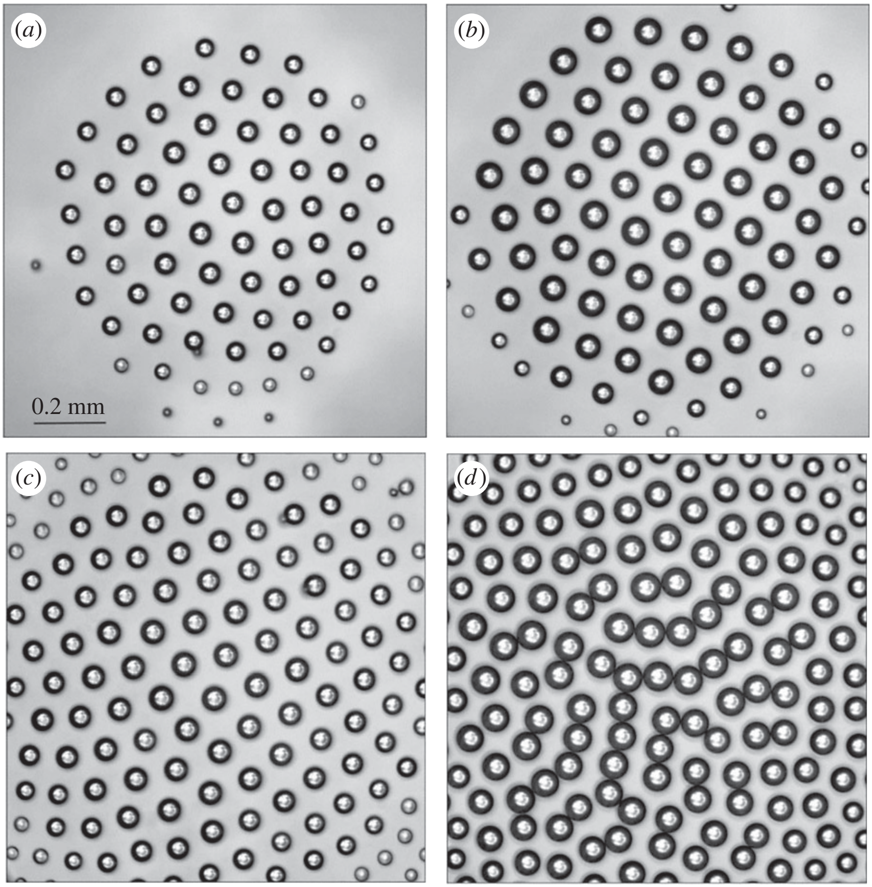

Figure 7. The evolution of droplet clusters with time of about 70 droplets (*a*) and (*b*) and of more than 200 droplets (*c*) and (*d*): (*a*) and (*c*) are the initial structures, (*b*) and (*d*) are the resulting structures. A large cluster ends up in the chain state, while a small one remains in the hexagonally ordered state.

produce sufficiently large self-assembled levitating clusters of water droplets which are similar to those observed at normal temperature conditions [36].

4. Transitions between hexagonally ordered and chain-like states

Another important recent observation is the possibility of a chain-like structure of a cluster instead of the usual hexagonal structure. The chain state with droplets almost touching each other and forming linear chains is found in the central part of the cluster, which is surrounded and 'compressed' by an external layer of droplets. The droplets should also be larger than a certain critical size for the transition from the hexagonally ordered to the chain state to occur. The transition from hexagonal to a chain cluster is reversible. It can be easily reversed by turning on infrared heating which suppresses the droplet growth due to condensation of water vapour. Two types of cluster evolution are shown in figure 7, with the only difference being in the initial size of the cluster (about 70 and more than 200 droplets). In a large cluster, central droplets exceed a certain critical diameter and a chain cluster is formed.

Note that while the hexagonal cluster is more ordered than the chain cluster (as follows from the analysis of the Voronoi entropy of the cluster), the latter is also not completely disordered or chaotic. While this transition between the two states is somewhat similar to the second-order phase transition between an ordered and disordered state, for example, in dusty plasma [37],

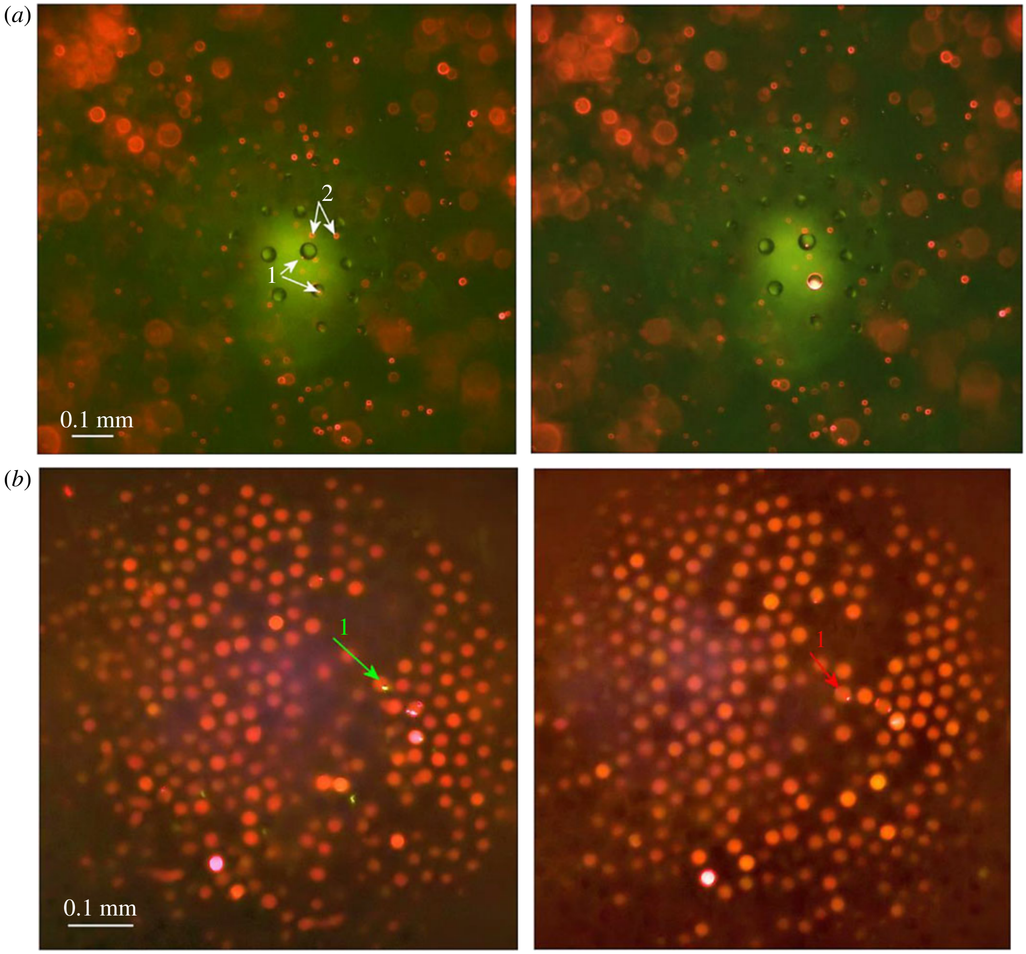

Figure 8. Microorganisms encapsulated into a droplet cluster: (*a*) *Chlorella vulgaris Beijer*, chlorophyll fluorescence, time interval 2 s between the frames; (*b*) *Escherichia coli* K12, coloured with LIVE/DEAD BacLight Bacterial Viability Kit (Thermo Scientific, USA), 4 s between the frames. (Online version in colour.)

colloidal crystals, microfluidic dipoles [38] and droplet 'polymers' [39], the droplet cluster is a dissipative structure and, unlike the colloidal crystals, it does not exist at a phase equilibrium surface.

5. Biological implications of droplet clusters

Bacterial life and transport in microdroplets constitute an important topic which has not yet been studied in detail. Bioaerosols can contain bacteria and archaea, fungal spores and fragments, pollen, viruses, algae and cyanobacteria, biological crusts and lichens. Aerosolized microorganisms play a role in diverse processes ranging from ice and cloud nucleation to disease transmission and geographical migration of microbes [40]. Bioaerosols can be generated, for example, as a result of raindrops. Thus, a single raindrop can transfer 0.01% of bacteria on the soil surface and the bacteria can survive more than one hour after the aerosol generation process [41]. Metabolically active microbes have been detected in air at elevations as high as 20–70 km [40,42].

The main factor complicating the study of bioaerosols is that it is extremely difficult to observe airborne bacteria *in situ*, because it is impossible to trace individual microdroplets constituting aerosols. However, the droplet cluster technology allows the observation of isolated aerosol droplets for extended time spans (hours) and studying them with fluorescent microscopy.

We conducted a preliminary investigation of bioaerosols in stabilized droplet clusters with a fluorescent microscope Zeiss Axio Zoom V16 (light source HXP 200C, filter set 38 HE, camera pso.edge 5.5). Cells of a green microalgae *Chlorella vulgaris* and of non-pathogenic *Escherichia coli* bacteria, which are standard microorganisms for various microbiological experiments, were encapsulated into the droplets by ultrasound spreading. They can be observed in figure 8. The cells floating in the substrate layer can also be observed. While all cells have approximately the same size, those in the substrate look larger due to lens focusing on the droplets.

It is possible to distinguish between the living and dead cells by introducing a colouring matter into the droplets. Using a commercial staining kit containing DNA-intercalating dyes, we visualized treated bacterial cells with a permeable membrane. The lightweight green fluorescence SYTO 9 (approx. 400 Da) freely penetrates even through integral bacterial membrane; in contrast, heavier red fluorescence PI (668 Da) is accumulated inside only through disordered plasma membrane. Membrane disordering was spectroscopically recorded as quenching of the green fluorescence since the SYTO 9 is replaced by PI. The cells with undamaged membranes emit green light, while after breaking the cell membrane the colour changes to red.

The droplets rotate slowly, and the cells located on their surfaces can appear at the opposite from the lens side of the droplet. In this case, a bright flash is observed, since the droplet focuses fluorescent light radiation of the cell. Unfortunately, at this point in our preliminary experiments, the cells did not survive for longer than 4 s, with many of them dying already during the ultrasound spreading of the cells.

We consider these experiments as a preliminary step, which indicates that observation of microorganisms in a droplet cluster is possible in principle. In addition to the research of bioaerosols, droplet clusters can serve for chemical microreactors, in which droplet manipulation (e.g. merger of two droplets) combined with a chemical reaction can create interesting effects, such as implementation of logical computations [43].

6. Conclusion

In living nature, condensed microdroplets are often used for water collection by plants and insects in dry climates. Mechanisms of water harvesting often involve complex patterns in solid surfaces intended to capture condensed water. Artificial arrays of condensed microdroplets in breath figures and droplet clusters also involve patterned arrangements; however, these patterns are self-assembled. The breath figures are formed on smooth cold surfaces, while droplet clusters levitate over heated water in an ascending stream of air and vapour.

As most substances, water can exist in a solid, liquid and vapour state. The hexagonal symmetric structures are typical for the solid form of water, for example, snow crystals. However, both in breath figures and droplet clusters, a hexagonal arrangement is observed for liquid water. The interactions between droplets which control their equilibrium sizes and distances between them are quite complex and may involve the Marangoni flow in the case of the breath figures and complex aerodynamic forces in the case of the droplet cluster. Recent results demonstrate that electrostatic force is not involved in these interactions, and that the droplet cluster is essentially a 2D phenomenon (a monolayer of droplets). Whether 3D ordered microdroplet structures are possible, e.g. in clouds, cannot be concluded from droplet cluster observations.

We have presented new results on the reversible transition between the hexagonally ordered and recently discovered chain clusters. Microdroplets can also be used as biological microreactors. We also presented a feasible method to trace microorganisms in a cluster. While in principle individual microdroplets can be stabilized and traced for extended periods (hours), so far we have not yet been able to observe microorganisms living for extended periods (longer than seconds) in the droplets. Future *in situ* observation of airborne living cells in microdroplets would provide new insights on microorganism migration.

Data accessibility. This article has no additional data.

Authors' contributions. A.A.F. conducted the experiments with droplet clusters. E.B. studied breath figures. L.A.D. investigated the fluid mechanics of the droplet clusters and supplied figure 5. E.B., M.N. and L.A.D. studied the transitions between the hexagonal and linear clusters. M.N. supervised the writing of the manuscript. All authors read the final manuscript.

Competing interests. We declare we have no competing interests.

Funding. Supported by the Russian Science Foundation project no. 19-19-00076.

Acknowledgements. The authors are grateful to the Russian Science Foundation for the financial support of the present study. We are also grateful to Dr Alexey Vasilchenko for the samples of microorganisms used in the study.

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