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Bioaerosol Deposition on an Air-Conditioning Cooling Coil

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Abstract

This study is concerned with the role of a fin-and-tube heat exchanger in modifying microbial indoor air quality. Specifically, depositional losses of ambient bioaerosols and particles onto dry (not cooled) and wet (cool) coil surfaces were measured for different airspeeds passing through the test coil. Total, bacterial and fungal DNA concentrations in condensate water produced by a wet coil were also quantified by means of fluorescent dsDNA-binding dye and qPCR assays. Results revealed that the deposition of bioaerosols and total particles is substantial on coil surfaces, especially when wet and cool. The average deposition fraction was 0.14 for total DNA, 0.18 for bacterial DNA and 0.22 for fungal DNA on the dry coil, increasing to 0.51 for total DNA, 0.50 for bacterial DNA and 0.68 for fungal DNA on the wet coil. Overall, as expected, deposition fractions increased with increasing particle size and increasing airspeed. Deposited DNA was removed from the cooling coil surfaces through the flow of condensing water at a rate comparable to the rate of direct

deposition from air. A downward trend of bacterial and fungal DNA measured in condensate water over time provides suggestive evidence of biological growth on heat exchangers during nonoperational times of a ventilation system. This investigation provides new information about bioaerosol deposition onto a conventional fin-and-tube cooling coil, a potentially important factor influencing indoor exposure to microbial aerosols in air-conditioned buildings.

Key words: Bioaerosols, DNA, Deposition, Cooling coil, Condensate water

1. Introduction

The fin-and-tube heat exchanger is a ubiquitous component of air-conditioning systems in mechanically ventilated buildings, employed to condition the temperature and humidity of air delivered to indoor environments (Pongsoi et al., 2014; Tang et al., 2016). In warm seasons for temperate and subtropical climates, and during the whole year for tropical climates, the heat exchanger (commonly referred to as a cooling coil) is utilized to cool and dehumidify the air (Chen et al., 2016). The aggregate energy transfer at this location across all air-conditioned buildings accounts for much of the total energy demand and also peak energy demand during warm conditions in cities worldwide (Siegel and Carey, 2001).

The air supplied to mechanically ventilated buildings inevitably passes over heat-exchanger surfaces and the interaction between the air and the cooling coil surfaces can modify air quality, both for the flow path from outdoors to indoors and for recirculating airflows. Modeling and experimental studies reveal that some particles in the airstream could deposit onto the heat exchanger surfaces (Siegel and Nazaroff, 2003; Waring and Siegel, 2008; Gröhn et al., 2009; Grigonyte et al., 2014). Some studies also have suggested that previously deposited particles on coil surfaces could become reentrained in the airflow and constitute a secondary source of indoor particles (Siegel and Carey, 2001; Siegel, 2002).

Important knowledge gaps remain concerning how heat-exchanger surfaces in air-conditioning and mechanical ventilation (ACMV) systems influence indoor air quality. With regard to microbial air quality, it is important to note that heat exchanger surfaces are regularly wet in air-conditioning seasons in areas with moderate or elevated humidity. Observational studies have documented that the use of cooling coils could increase bioaerosol levels in indoor environments, suggesting a potential role of cooling coils as a source of indoor bioaerosols (Hugenholtz and Fuerst, 1992; Abe, 1998; Bluyssen et al., 2003; Jo and Lee, 2008). In addition to the potential release of biological materials, another process on cooling coil surfaces might also be important. Biological aerosol particles may deposit on cooling coil surfaces and be removed from air. Some fraction of the deposited particles may be transferred to the condensate water and be removed from the indoor environment through the drainage process. However, only one prior study has discussed the possibility of such bioaerosol deposition on cooling coil surfaces (Siegel and Walker, 2001). To the best of our knowledge, no work has yet been published that experimentally investigates bioaerosol deposition processes onto an air-conditioning cooling coil.

The objective of this research is to provide a systematic experimental investigation of bioaerosol transformations across a typical fin-and-tube heat exchanger in a model vapor-compression ACMV system similar to those used in modern air-conditioned buildings in tropical environments. In brief, the deposition fractions of ambient bioaerosols that include bacterial and fungal aerosol particles, size-resolved total particles, as well as monodisperse polystyrene latex (PSL) particles were measured for dry (not cooled) and wet (cooled) coils. Total, bacterial and fungal DNA concentrations in condensate water draining from a wet coil were also analyzed using a Qubit fluorometer and real-time PCR system. This study contributes to a better understanding of bioaerosol transformation processes as pertinent influences of indoor microbial air quality in air-conditioned buildings.

2. Materials and Methods

2.1. Experimental apparatus and test procedure

This work was conducted using a laboratory apparatus (see Figure 1) in which a fin-and-tube cooling coil system was situated between connecting upstream and downstream ducts. The cooling coil was of conventional design, comprising four rows of cylindrical copper refrigerant tubes, which were oriented horizontally and to which were attached vertical aluminum fins. The apparatus had a fin pitch of 3.1 fins/cm (within the common range of 2.4 to 7.1 fins/cm) and a center-to-center tube spacing of 7.6 cm. The corrugated fins were 0.1 mm thick and 44 mm deep in the direction of air flow. The copper tubes, which were inserted into aluminum vertical fins with full fin collars, had an outer diameter of 1.59 cm and a 0.09 cm thick wall.

The test apparatus was sited in a laboratory that was open to ambient air during working hours. A variable speed fan, installed at the inlet of the test coil system, pushed air through 3.6 m of straight 42 cm × 42 cm square upstream duct. The air then passed through the fin-and-tube heat exchanger which has the same area as the upstream duct (42 cm × 42 cm), followed by another 3.6 m section of straight 42 cm × 42 cm square duct downstream. Air speeds inside air-handling unit cooling coil systems commonly range from 1 to 4 m/s (Siegel and Nazaroff, 2003; Siegel and Carey, 2001). In this work, we used the variable speed fan to test three air speeds for the open sections of the ducts: 1.0, 1.5 and 2 m/s. If not otherwise specified, the results presented here are for an air speed of 1.5 m/s, which converts to a mass flow rate of 1030 kg/h of dry air passing through the coil surfaces (Rim et al., 2015).

This work was conducted in Singapore, where the ambient dew point temperature is consistently high. Dry-bulb temperatures typically range from 25 to 32 °C and the ambient relative humidity (RH) was always above 75% during these experiments. Continuous measurements were made of air temperatures, RH and air speeds using air velocity meters

(VELOCICALC Air Velocity Meter Model 9545, TSI Inc., Shoreview, MN, USA) at two cross-sections that were positioned 0.5 m upstream and 0.5 m downstream of the cooling coil. We utilized nine measurement locations defining a uniform grid through each of the two cross-sections of the duct. Airflow parameter values are reported in Figure S1 in the supporting information. When the cooling coil was operated, surface temperatures were also continually measured using digital thermometers (Fluke 54 II B Dual Input Digital Thermometer, Fluke Corporation, WA, USA). The measurement points were located on the top of the side face for each of the four rows of copper refrigerant tubes (as marked by red triangles in Figure 1). Results are shown in Figure S2.

In this work we studied two operational conditions: cooling coil off ("dry coil," which refers to a coil that is not being cooled and that is also not wet) and cooling coil on ("wet coil," which refers to a cooled coil onto which condensation occurs continuously). For the dry coil, the fan was on and the compressor was off for all experimental time. The entire system was dry and nominally isothermal. Conversely, for the wet coil, the fan and the compressor were on at all times. Cooling was achieved, and condensate water was produced, by sending the coolant R-134a through the cooling coil system. Both modes of operation were tested in steady state with regard to thermal conditions.

In our experiments, the surface temperatures of the wet coil (16 - 6 °C from Row 1 to Row 4, see Figure S2) were much lower than those of dry coil (room temperature, ~ 26 °C). For all experiments, the cooling system as well as the fan were operated from 8 AM to 7 PM, Monday through Friday, and were off for other times to simulate one type of typical operation in commercial buildings in Singapore (Rim et al., 2015). In this work, we found the wet coil could produce the condensate water at a relatively stable flow rate after the

compressor was on for an hour. So, for the wet coil condition, all measurements were conducted starting from 9 AM, i.e. after 60 min of coil operation.

2.2. Leakage test of the coil system

Before conducting the experiments, we cleaned the cooling coil with freshly purified water and checked the test system for cleanliness and for leakage. We installed a HEPA filter at the inlet of the system to remove particles from the ambient air and monitored the particle concentrations both for upstream and downstream of the dry coil using optical particle counters (OPC, AeroTrak® Handheld Particle Counter Model 9306, TSI Inc., Shoreview, MN, USA). The ambient particle concentrations were also monitored at the same time using three OPCs. Results confirmed that the maximum leakage rate was small enough to not significantly influence the study objectives, as shown in Figure S3.

2.3. Particle concentration monitoring

2.3.1 Total particles

Size-resolved particle concentrations were monitored upstream and downstream of the cooling coil with OPCs and scanning mobility particle sizers (NanoScan SMPS nanoparticle sizer 3910, TSI Inc., Shoreview, MN, USA) for both dry and wet coil conditions. The upstream and downstream sampling arrays were located in the cross-sectional area of the duct 2.6 m upstream and 2.6 m downstream of the test coil, respectively. Each sampling array comprised three equidistant vertical columns of Teflon sampling tubes into which were drilled four equally spaced holes. The inner diameter of holes was selected based on the air speed inside the duct to approximately achieve isokinetic sampling. The OPC measures particle number concentrations in six size ranges: 0.3-0.5 μm, 0.5-1.0 μm, 1.0-2.5 μm, 2.5-5 μm, 5-10 μm and 10-25 μm. The SMPS was measured size-resolved particle concentrations spanning the diameter range 10 to 420 nm.

2.3.2. Monodisperse polystyrene latex (PSL) particles

To investigate in more detail the size-dependence of particle deposition on the cooling coil, we also measured deposition using monodisperse spherical PSL particles (JSR Trading Co., Ltd., Tokyo, Japan) across both the dry and wet coil. The PSL suspension (concentration = 10^8 particles/mL) was aerosolized using a Collison nebulizer (CN24, BGI Inc., Waltham, MA), which was operated at a flow rate of 2.5 L/min with nitrogen gas. The resulting aerosol flow was delivered into the upstream zone of the test system. During the PSL tests, a HEPA filter was installed at the duct inlet to remove ambient particles from the test system. Airborne PSL concentrations were measured upstream and downstream of the coil with OPCs in the same locations in the duct as total particles described in §2.3.1. Four PSL diameters were tested independently: 0.8, 2.0, 3.3 and 5.1 μm. Nine replicate experiments were conducted for each size.

2.4. Bioaerosol sampling

To collect bioaerosol samples for subsequent analysis, during each experimental trial, air upstream and downstream of the coil was isokinetically sampled for 9-h periods (daily from 9 AM to 6 PM) using in-line sterilized membrane filters (Super 200, Pall Corporation, Michigan, USA) at a sampling flow rate of 15 L/min with similar sampling arrays as for particles (see §2.3.1). The air sampling flow rate was calibrated using a Gilibrator-2 (Sensidyne, Inc., Petersburg, FL, USA) both before and after sampling. We also placed a quality-control blank filter in a holder and left it exposed in the ambient air near the test coil system for 9 hours.

We compared the in-line sampling method with open-face filter sampling (upstream of the coil) to check the possibility that some of the bioaerosol DNA might have deposited on the sampling tube before reaching the filter. In the verification test, the middle of the three Teflon sampling tubes was removed and replaced by two open-face filter holders, which were

182 directly located in the center of cross-sectional area of the duct. The two in-line sampling
183 filters and the two open-face filters were simultaneously operated with their pumps to collect
184 bioaerosols at a standard flow rate of 15 L/min for 9 hours. All bioaerosol samples were
185 analyzed with the molecular methods described in §2.6. Results of this verification test
186 showed good comparability between the open-face and in-line samplers, as presented in
187 Table S1.

188 *2.5. Collection of condensate water produced by wet AC cooling coil*

189 Condensate water samples of 1-L volume were collected at intervals of 60 min (collection
190 times = 9 AM through 2 PM) using sterile centrifuge tubes (Corning, Inc., New York, USA)
191 for all experiments in which the coil was cooled. In addition, 500-mL samples of condensate
192 water from an AHU air-conditioning system used in a commercial building in Singapore
193 were also collected at 8 AM, 2 PM and 6 PM on Fridays for three successive weeks. In
194 contrast to the laboratory test system, the commercial-building AHU processed return air
195 from offices that had high human occupancy. All water samples were analyzed with the
196 molecular methods described in §2.6.

197 *2.6. DNA extraction and quantification*

198 After sampling, DNA was extracted from membrane filters used for air sampling and
199 from the blank control filters. Condensate water was first processed using filter funnels with
200 0.22 μm membrane filters (MO BIO Laboratories, Carlsbad, CA, USA) and then these filters
201 were extracted for DNA using the same procedure as for air-sampling filters. Extractions
202 were performed using the PowerWater® DNA Isolation Kit (MO BIO Laboratories, Carlsbad,
203 CA, USA). We followed the manufacturer's recommended procedures with an additional step
204 of water bath sonication to improve DNA yield (Luhung et al., 2015). To briefly summarize,
205 first, the filter was placed into a 5-ml tube with 1 ml preheated PW1 (a strong lysing reagent),

followed by incubation in a 65 °C sonication water bath for 30 min and centrifugation at 4000 × g for 1 minute. Avoiding the pellet at the bottom of the tube, the supernatant was transferred to a clean 2-ml collection tube. Second, 200 µl of PW2 solution was added to the suspension to remove non-DNA organic and inorganic materials, including humic acid, cell debris, and proteins. After incubating at 4 °C for 5 min, samples were centrifuged again at 13,000 × g for 1 min. The remaining extraction steps were performed according to the standard MO-BIO PowerWater® DNA isolation protocol. Finally, 60 µL of solution from each sample was saved for subsequent DNA quantification and amplification. All manipulations of the samples were performed in a Biosafety Level II cabinet (Sterilchem GARD, Baker Co., Sanford, Maine).

DNA concentrations (both microbial and non-microbial) were quantified by means of a fluorescent dsDNA-binding dye assay (Qubit Fluorometer, Invitrogen, Carlsbad, CA, USA) according to manufacturer's procedures. Concentrations of DNA extracted from blank samples were consistently below the detection limit of Qubit fluorometer, which indicates that there was no significant DNA contributed by laboratory personnel.

In addition to total DNA quantification, the concentrations of bacterial and fungal DNA were also quantified using qPCR. Bacterial universal forward primer 5'-TCCTACGGGAGGCAGCAGT-3' (T_m, 59.4 °C), reverse primer 5'-GGACTACCAGGGTATCTAATCCTGTT-3' (T_m, 58.1 °C) and probe (6-FAM)-5'-CGTATTACCGCGGCTGCTGGCAC-3'-(TAMRA) (T_m, 69.9 °C) were used for bacterial DNA amplification (Nadkarni et al., 2002). Universal forward primer 5'-GGRAAACTCACCAGGTCCAG-3' FungiQuant-F (T_m, 62.5 °C), reverse primer 5'-GSWCTATCCCCAKCACGA-3' FungiQuant-R (T_m, 56.5 °C), and probe (6FAM)5'-TGGTGCATGGCCGTT-3'(MGBNFQ) (T_m, 68 °C), designed by Liu et al. (2012), were used for fungal DNA amplification. The amplification and detection of genes was performed

using Applied Biosystems Step-one real-time PCR system and the qPCR conditions described by Liu et al. (2012) were used in this study.

DNA extracted from *Escherichia coli* (ATCC 15597) and *Aspergillus versicolor* (ATCC 26644) suspensions were serially diluted into 10^{-1}, 10^{-2}, 10^{-3}, 10^{-4} and 10^{-5}, respectively. These dilutions were then quantified with the Qubit fluorometer and used as bacterial and fungal DNA standards for qPCR. The bacterial and fungal suspensions that we used were separately prepared following procedures described in previous studies (Wu and Yao, 2010; Liang et al., 2012).

2.7. Quantifying bioaerosol and particle deposition fractions

The deposition fraction of total particles, PSL and bioaerosol DNA on the cooling coil, η, was evaluated using this equation:

$$\eta = [1 - (C_{downstream} / C_{upstream})] \quad (1)$$

where $C_{upstream}$ is the analyte concentration of the air upstream and $C_{downstream}$ is the analyte concentration of the air downstream of the cooling coil. Bioaerosol deposition fractions were calculated using measured DNA concentrations from the sampling filters. The deposition fraction for particles and PSL was determined from measured number concentrations.

2.8. Quantifying condensate generation rate and calculated DNA concentration in condensate water

The generation rate of condensate water depends on environmental conditions and can be calculated from air temperature, relative humidity and air flow rate data, as shown in equation (2):

$$Q_{condensate} = F_{dryair} \times (W_{upstream} - W_{downstream}) / \rho \quad (2)$$

In this equation, $Q_{condensate}$ is the volumetric flow rate of condensate water from the cooling coil, under steady drainage conditions (L/min), F_{dryair} is mass flow rate of dry air through the

coil (kg min^{-1}), W is the humidity ratio of the air upstream and downstream of the coil (kg of water vapor per kg of dry air), and ρ is the density of liquid water (1.0 kg liquid water per L). The humidity ratio is calculated using equation (3):

$$W = 0.622\, P_w / (P - P_w) \quad (3)$$

where P_w is the partial pressure of water vapor, evaluated from the relative humidity and air temperature (see, e.g., Table 1 in Rim et al., 2015) and P is the total air pressure.

We computed a benchmark estimate of the DNA concentration in condensate water by assuming that the DNA lost from the airstream was transferred quantitatively to the draining condensate. The measured concentration could be compared to this benchmark to provide an indication of whether (a) much of the deposited DNA was retained on the coil or (b) DNA was shed as an outcome of microbial replication on coil surfaces. The benchmark estimate is computed from equation (4):

$$C_{benchmark} = F_{dryair} \times (C_{upstream} - C_{downstream}) / Q_{condensate} \quad (4)$$

Here, $C_{benchmark}$ is the predicted DNA concentration in the condensate water (mass of DNA per L) based on the assumption of no loss or gain of DNA owing to interactions with cooling coil surfaces, $C_{upstream}$ is the DNA concentration of the air upstream and $C_{downstream}$ is the DNA concentration of the air downstream of the cooling coil, both expressed as mass of DNA per mass of dry air.

2.9. Statistical Analysis

The experimental data were analyzed by a paired *t*-test utilizing SigmaPlot version 10 software, and one-way ANOVA tests. Values of $p < 0.05$ were taken to indicate a statistically significant difference.

3. Results

3.1. Particle deposition fraction

As expected, the deposition fraction on the test coil varied with particle size. For supermicron particles, the deposition fraction increased with increasing particle size for both dry and wet coils (see Figure 2). Across the diameter range 1.0 to 25 μm, the deposition fractions of particles were found to be higher than 0.11 (Figure 2(a)), with a maximum value of 0.65 for the wet coil and 0.50 for the dry coil. For smaller particles, in the diameter range 30 nm to 1.0 μm, deposition fractions were found to be small and not strongly dependent on particle size (Figure 2(a) and Figure S4). The monodispersed PSL particle deposition was also found to increase with increasing particle size, as shown in Figure 2(b). Increasing deposition rate with increasing size for supermicron particles is consistent with expectations, with inertial drift being the dominant particle transport process inducing deposition (Siegel and Nazaroff, 2003).

We observed that the deposition fraction was substantially higher onto wet coil surfaces than onto dry coils for supermicron particles, both for total ambient particles and for monodisperse PSL particles. Note that the four rows of fin-and-tube coil surfaces were dry and isothermal for the dry coil condition, but were wet with a continuous flow of condensing water for the wet coil condition. The tube surfaces for the wet coil were progressively cooler in the streamwise direction, ranging from 16 °C for Row 1 to 6 °C for Row 4 (Figure S2).

Two mechanisms could contribute to the higher deposition fraction on wet coil surfaces. First, condensed moisture on the cooling coil surfaces would produce a decreased channel width, inducing higher air speeds and lower travel distances, and thereby enhancing inertial impaction. A second possible contributor to enhanced particle deposition is faster transport toward the surface owing to one or both of thermophoresis, induced by the thermal gradient toward the cool coil surfaces, and diffusiophoresis, induced by the net flux of water vapor

toward the condensing coil surface.

3.2. Bioaerosol deposition fraction

We found substantial bioaerosol particle deposition to the cooling coil, especially when cool and wet. Downstream air was found consistently to have lower DNA concentrations in bioaerosols than upstream air both for dry and wet coils (Figures 3 and 4). For the dry coil, the deposition fractions were small: 0.14 for total DNA, 0.18 for bacterial DNA and 0.22 for fungal DNA, respectively. By contrast, when the coil surfaces were cool and wet, with active water condensation from the air, the deposition fractions increased markedly: 0.51 for total DNA, 0.50 for bacterial DNA and 0.68 for fungal DNA, respectively. For the dry coils, the measured deposition fractions were not statistically different from zero (p = 0.25 - 0.38). However, for the wet coils, total, bacterial, and fungal DNA concentrations differed between the upstream and downstream sampling locations with statistical significance (p-values < 0.05).

The cooling coil exhibited similar removal effects for bacterial and fungal bioaerosol particles (Figure 4), although there was somewhat higher deposition for fungal DNA than for bacterial DNA. These measured differences might have been originated from different particle sizes. Fungal bioaerosols may be larger than particles containing bacterial DNA (Després et al., 2012), and, as we have shown for particles (Figure 2), larger particles exhibit a larger deposition fraction than do smaller particles in the supermicron size range.

Considering the data in Figures 3 and 4, we also found that the total bacterial and fungal DNA (71-355 fg of DNA per kg of dry air) contributed only a very small fraction of total DNA (64-70 pg of DNA per kg of dry air) to the air samples. In addition to microbial DNA, environmental DNA-containing bioaerosols can include pollen, viral DNA, vegetation debris and insect debris (Eduard et al., 2012).

We observed that the deposition of bioaerosols could also be affected by air speed

through the test coil. Although the general patterns of fractional DNA deposition across the cooling coil were similar, the deposition fractions for air speeds of 1, 1.5 and 2 m/s were distinguishable (Figures 3 and S5). For the dry coil, the deposition fraction of total bioaerosol DNA increased from 0.11 for an air speed of 1 m/s to 0.16 for 2 m/s. On the cool, wet coil, the deposition fraction of total DNA increased from 0.44 for an air speed of 1 m/s to 0.54 for 2 m/s. Thus, higher air speeds produced more deposition for both dry and wet coils, indicating that air speed is another parameter that affects biological particle deposition onto cooling coil surfaces.

From the data presented in Figures 3 and 4, one can infer that 36 pg of total DNA (70 upstream vs. 34 downstream pg of DNA per kg of dry air), 189 fg of fungal DNA (278 upstream vs. 89 downstream fg of DNA per kg of dry air), and 45 fg of bacterial DNA (90 upstream vs. 45 downstream fg of DNA per kg of dry air) would have been deposited onto wet cooling coil for every kg dry air passing through the cooling coil. Based on previous studies, 45 fg of bacterial DNA, for example, could correspond to approximately 9 bacterial cells if using *Escherichia coli* as a standard (Nadkarni et al., 2002; Raghunathan et al., 2005). As the cooling system was operated from 8 AM to 7 PM, one can estimate that the maximum daily deposition for bacterial particles onto the tested cooling coil was 0.1 million bacterial cells. If some proportion of these bacterial cells remain attached, remain viable and reproduce, then some of the generated cell mass and/or the microbial metabolic byproducts might be re-entrained into the air stream, potentially degrading indoor air quality.

3.3. DNA collected in condensate water

Figure 5 displays a time-series of DNA concentrations — total, bacterial, and fungal — from condensate water produced by the wet coil. Total DNA concentrations (10-13 ng/L) were much higher than bacterial (0.04-0.2 ng/L) and fungal (0.01-0.3 ng/L) concentrations. Both bacterial and fungal DNA contributed less than about 1% of total DNA in the

condensate water, similar to the airborne proportion. We also observed that bacterial DNA concentrations in the condensate water were much higher than the fungal concentrations, except for the first hour of the operation. The relative abundance of bacterial and fungal DNA in condensate water was different than their relative concentrations in the air (278 fg of fungal DNA vs. 90 fg of bacterial DNA per kg of dry air) as well as the respective reductions from the airstream (189 fg of fungal DNA vs. 45 fg of bacterial DNA reduction per kg of dry air) as shown in Figure 4. Due to the fact that bacteria excel in replication and have much faster reproduction rates than fungi (Coleman, 1994; Kirchman, 2012), we speculate that a possible reason for the observation is that some previously deposited bacteria remained viable and might have reproduced on the coil surfaces, which then resulted in elevated bacterial DNA concentrations as compared with fungal DNA concentrations in the condensate water.

Total DNA and microbial DNA concentrations in the condensate water exhibited different time trends. The total DNA concentrations were fairly stable (ranging from 10 to 13 ng/L) throughout the day. That result is consistent with a hypothesis that this DNA was in condensate water directly as a consequence of current deposition from air onto coil surfaces followed by an effective washing of the deposited DNA with the condensing water. In contrast, both bacterial and fungal DNA concentrations showed a downward trend with time during the course of a sampling day. This aspect was especially distinctive early: the concentration of microbial DNA in condensate water for the first hour of operation was an order of magnitude higher than the concentrations in later samples. A similar downward trend was also found for bacterial DNA concentrations in the condensate water produced by cooling coils from a commercial-building AHU (Figure S6).

A plausible explanation for this observation is that the microbial DNA was systematically higher in ambient air during the morning hours. However, it seems improbable that the

difference in airborne microbial DNA levels with time of day would be sufficiently large to explain the condensate-water trends. An alternative possible explanation is that some of the deposited bacteria and fungi were viable and the viable microbes may have reproduced on cooling coil surfaces while the system was off overnight. It is feasible that viable bacteria and fungi deposited during the daytime grow on cooling coil surfaces during the night if environmental conditions (e.g., temperature, moisture, and nutrient levels) are suitable. Biodegradable organic matter from both the gas and particle phase could deposit on the cooling coil during operation providing a substrate for microbial growth. Considering the moist surfaces during operation, the persistently high RH and the chronically warm temperatures in Singapore, cooling coil surfaces might provide very good conditions for microbial growth during daily and/or weekly periods of ACMV system inoperation. A recent study in Singapore found evidence of rapid growth of *Sphingomonas* on a detergent-cleaned cooling coil surface, with a relative abundance of 5% in the first 3 days after the cleaning to a relative abundance of 30% in 11th days after the cleaning, while its relative abundance in the upstream air was always smaller than 5% (Acerbi et al., 2016).

3.4. Measured and calculated DNA concentrations in condensate water

Figure 6(a) compares measured and computed DNA concentrations in the condensate water for the first consecutive seven days of operation in the wet mode. The computed concentration ranged from 24 to 43 ng/L. The measured concentration was more variable, ranging from 10 to 63 ng/L, with a maximum value four times higher than the central tendency values shown in Figure 5. For the first four days, measured concentrations in the condensate water were elevated above the computed values. These elevated DNA collection rates at the early stage of wet coil operation might be explained by the measured values including not only the currently deposited materials from the air, but also contributions from the washing of previously accumulated DNA, e.g., from dry coil operation. Starting from the

fifth day of wet coil operation, measured DNA concentrations in the condensate water began to be lower than the calculated values, suggesting that some newly depositing bioaerosols materials weren't being removed and remained in the test coil system. The difference between the measured and calculated concentrations of DNA in condensate water shows a steady downward trend (Figure 6(b)).

Interpreting the combined the data depicted in Figures 5 and 6, it appears that the condensate water serves a cleaning function for the cooling coil system, removing a large portion of the deposited biological materials from the cooling coil surfaces. However, it also appears that some biological materials that deposit from the air remain in the cooling coil system, at least temporarily. If some of the retained DNA is associated with viable microbes, they could contribute to the fouling of heat exchanger surfaces, diminishing the designed purpose of the heat exchanger and also potentially degrading indoor air quality.

4. Discussion

4.1. Deposition fraction comparisons with literature

Data regarding particle deposition on the test coil can be compared with previous modeling and experimental studies (Siegel and Carey, 2001; Siegel, 2002; Siegel and Nazaroff, 2003; Waring and Siegel, 2008). For example, Siegel and Nazaroff (2003) reported that particle deposition on cooling coils should increase with increasing diameter and increasing airspeed based on model predictions and experimental measurements, outcomes that are qualitatively similar to the findings in the present work. The predicted deposition fractions from that study are somewhat smaller than measured in the present study. Contributing to the difference may be the geometries of the two heat exchangers as well as the climatic factors in which these two studies were conducted. Based on modeling of transport and deposition processes, previous studies also suggest that deposition of supermicron particles should be enhanced to a wet cool coil surface as compared to a dry

isothermal surface (Siegel, 2002; Waring and Siegel, 2008), a prediction that is qualitatively consistent with our experimental data, as shown in Figure 2.

The present study contributes new experimental data regarding bioaerosol deposition onto cooling coils. Although there is a literature discussing total particle deposition based on either modelling or experiments, there are no prior published studies that have directly assessed deposition of bioaerosol particles. Based on a mass-transport model, Siegel (2002) suggested that bioaerosols would have deposition fractions in the range 20-40% for cool, condensing coils with an air speed of 1.5 m/s. Here, we have obtained similar but somewhat higher experimental values. Because of the importance of particle size as an influencing factor, we anticipate that the DNA deposition fraction would vary in accordance with the varying bioaerosol size distributions in the airstream approaching the cooling coil. However, it remains a considerable challenge to measure the particle size distribution of microbial DNA in air (e.g., Yamamoto et al., 2014). In the absence of size-resolved data, the aggregate measures we report here may be useful as a basis for quantitative assessments.

4.2. Microbial growth on cooling coil surfaces

Previous studies have hypothesized that the moist areas on and around cooling coils may provide suitable conditions for microbial growth (Hugenholtz and Fuerst, 1992; Muyshondt et al., 1998; Levetin et al., 2001; Schmidt et al., 2012). Some studies have reported that the use of air-conditioners was associated with elevated levels of microorganisms in the indoor spaces, which was observed immediately after switching on the air conditioning (Abe, 1998; Hamada and Fujita, 2002; Jo and Lee, 2008). From the present work, the downward trend of bacterial and fungal DNA measured in condensate water over time (Figure 5) provides new experimental information supporting this hypothesis. In addition, the finding that the concentration in condensate water for the first hour was an order of magnitude higher than the concentrations at later times suggests the possibility of overnight reproduction of viable

bacteria and fungi on the cooling coil surfaces.

4.3. Is bioaerosol deposition on cooling coil surfaces good or bad?

We found that deposition of particles and bioaerosols was substantial onto cooling coil surfaces. Cooling coil surfaces could contribute to the removal of particles and bioaerosols from the airflow, thereby contributing to lower airborne concentrations and associated human exposures in occupied spaces. Viewed narrowly, deposition on cooling coil might be considered a good outcome. However, particle and bioaerosol retention on cooling coil surfaces may also result in subsequent problems. One concern is increased energy requirements owing to the addition of thermal resistance associated with deposited materials and/or biofilms forming on the fin-and-tube heat exchanger, as has been suggested in previous studies (Siegel and Carey, 2001; Waring and Siegel, 2008; Wilson et al., 2013; Cremaschi and Wu, 2015). A second concern is reaerosolization of deposited materials and (potentially) the release of metabolic byproducts of microbes along with spores and fragments. This second concern, in which the cooling coil surface becomes a potential site for net bioaerosol emissions, was indicated by the work of Hugenholtz and Fuerst (1992). They found that post-coil air had much higher bacterial concentrations than pre-coil air. A third concern is the possible degradation of cooling coil surfaces caused by the deposited materials, which may contribute to a shortening of the useful life of these components in air conditioning systems.

5. Conclusions

People in indoor environments commonly inhale air that has contacted fin-and-tube heat exchangers. The interaction between the air and the cooling coils can influence indoor air quality. One important concern is bioaerosols. This investigation has provided new quantitative information regarding how fin-and-tube heat exchangers modify indoor

bioaerosol air quality. We have primarily reported on the experimental deposition fractions of bioaerosols measured across fin-and-tube cooling coils in a laboratory model of a core portion of an air handling system. In this study, we found that the cooling coil is a substantial sink for bioaerosols and exhibits similar deposition patterns for bacterial and fungal bioaerosol particles. The fractional particle deposition depends on particle size, air speed and operation mode of the cooling coil. We have also found that the flow of condensing water from wet coils carries DNA at rates comparable to the rates of direct deposition from air. The decreasing trend of bacterial and fungal DNA concentrations in the condensate water provided suggestive evidence for microbial growth on heat exchanger surfaces during overnight periods when the ventilation system was not operated. Comparing the measured DNA concentrations in condensate water with calculated values based on DNA deposition from the airstream, we also found evidence that some fraction of biological particles remained on cooling coil surfaces, which may cause subsequent fouling and may also be a source for indoor bioaerosols and their metabolic byproducts. These new data and the interpreted insights are relevant to the hygienic design and operation of ACMV systems.

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Appendix: Supplementary Information

Supplementary information may be found in the online version of this article:

499 Figure S1. (a) Temperature, (b) relative humidity and (c) air speed of the upstream and
500 downstream air with a wet cooling coil when the air speed was 1.5 m/s.
501 Figure S2. Time-resolved surface temperatures of four rows of cylindrical copper refrigerant
502 tubes with a wet coil when the air speed was 1.5 m/s.
503 Figure S3. Size-resolved particle concentrations of the ambient air, upstream and downstream
504 of the coil for the leakage test.
505 Figure S4. Deposition fractions of particles in the diameter range of 10 - 205 nm obtained by
506 SMPS.
507 Figure S5. The concentrations of DNA in bioaerosols sampled from air upstream and
508 downstream of the cooling coil when the air speeds were (a) 1 m/s and (b) 2 m/s.
509 Figure S6. Bacterial DNA concentration in condensate water collected at different times for
510 three consecutive weeks from an air handling unit in a commercial building.
511 Table S1. Comparisons between in-line and open-face sampling methods for bioaerosols.

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Figures

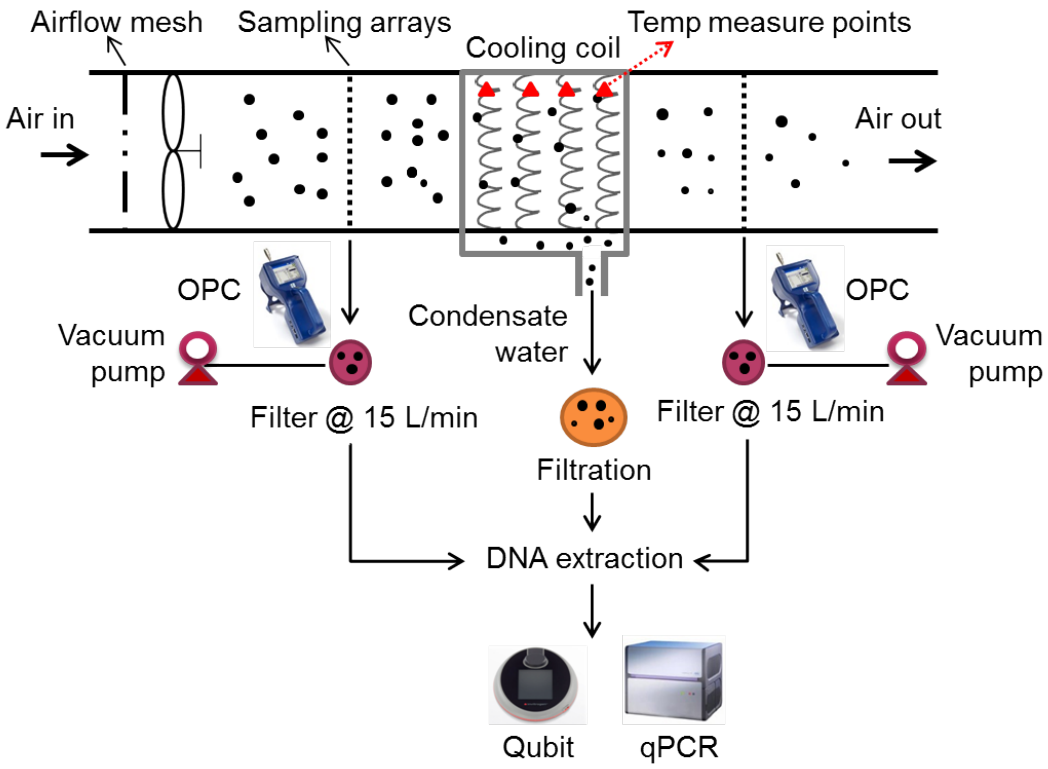

Figure 1. Schematic diagram of system for studying particle and bioaerosol deposition onto an air-conditioning cooling coil.

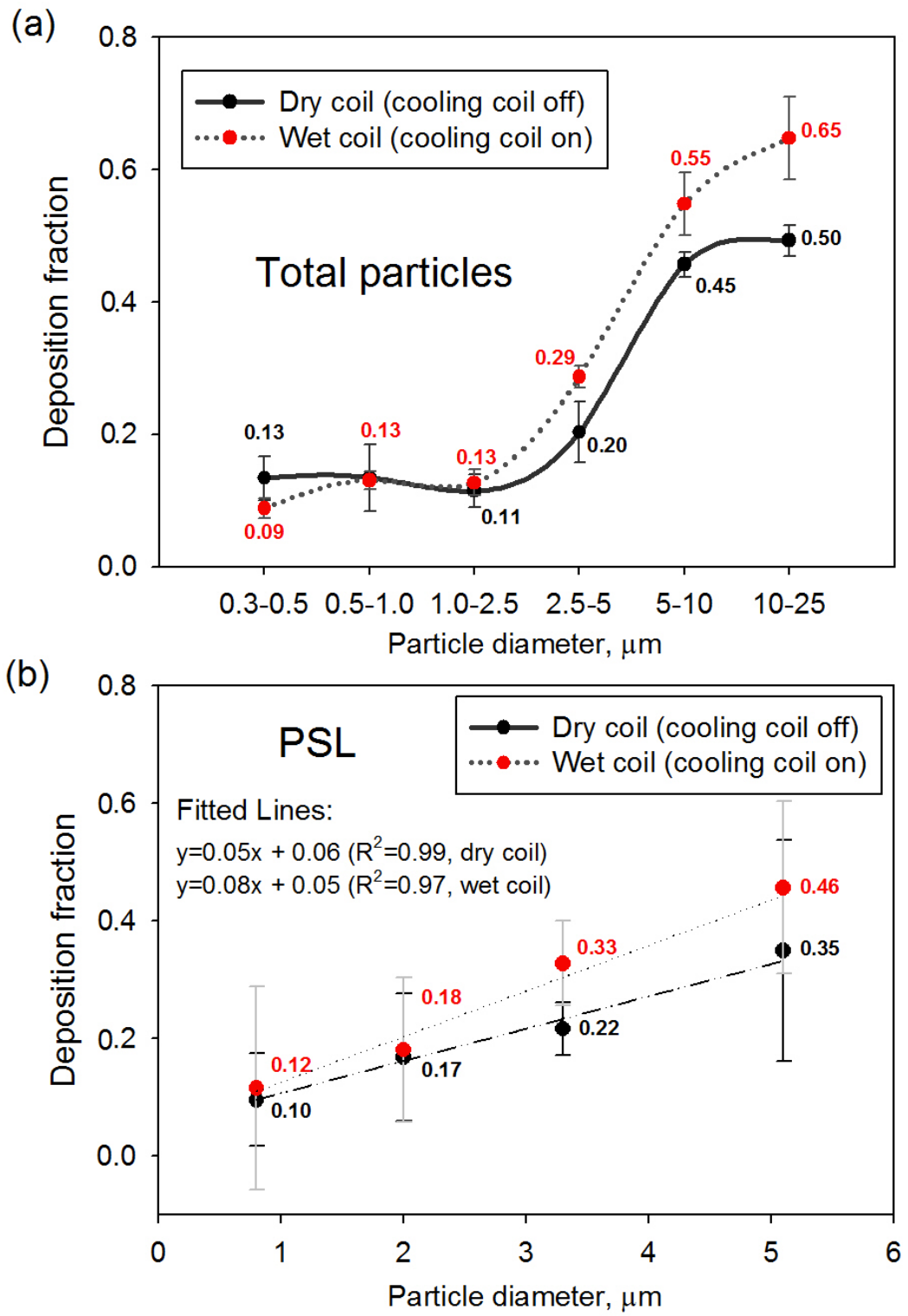

Figure 2. Deposition fractions of (a) total size-resolved particles and (b) monodisperse polystyrene latex (PSL) particles (diameters = 0.8, 2.0, 3.3 and 5.1 µm) onto the cooling coil when the air speed was 1.5 m/s. In (b), lines indicate the best-fit linear correlation between deposition fraction (y) and particle diameter (x, µm). Data points represent averages of nine independent repeated experiments and error bars represent the standard deviations. Numbers labeling the points indicate the mean particle deposition fractions under different cooling coil operation conditions.

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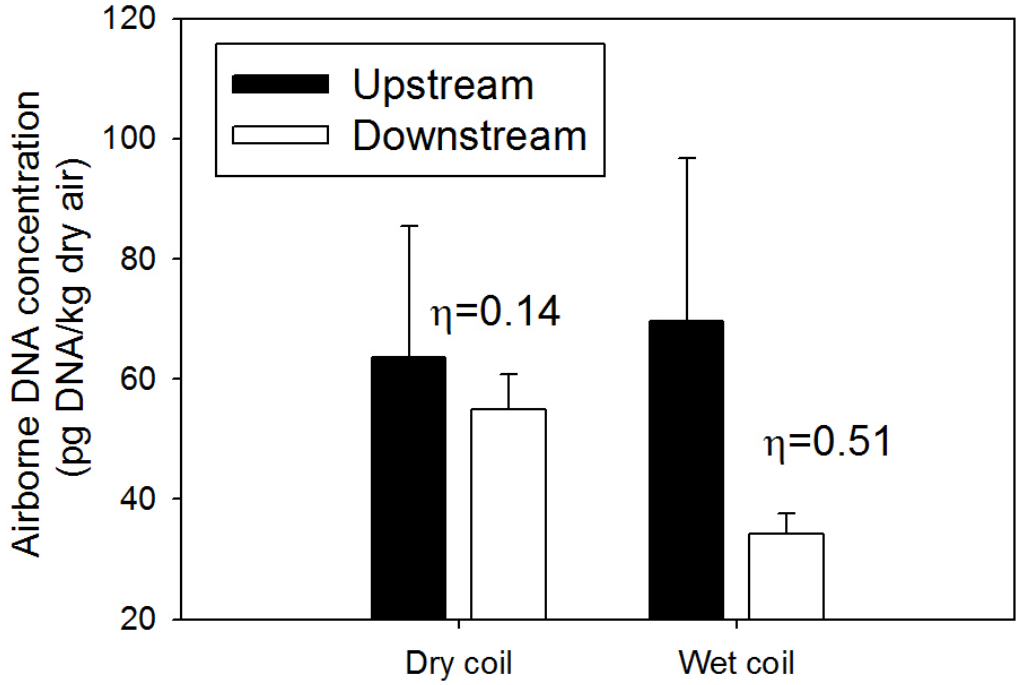

619

620 **Figure 3.** The concentrations of DNA in bioaerosols sampled from air upstream and
621 downstream of the cooling coil when the air speed was 1.5 m/s. The symbol η represents the
622 DNA deposition fraction on the cooling coil. Data points represent averages of nine
623 independent repeated experiments and error bars represent the standard deviations.

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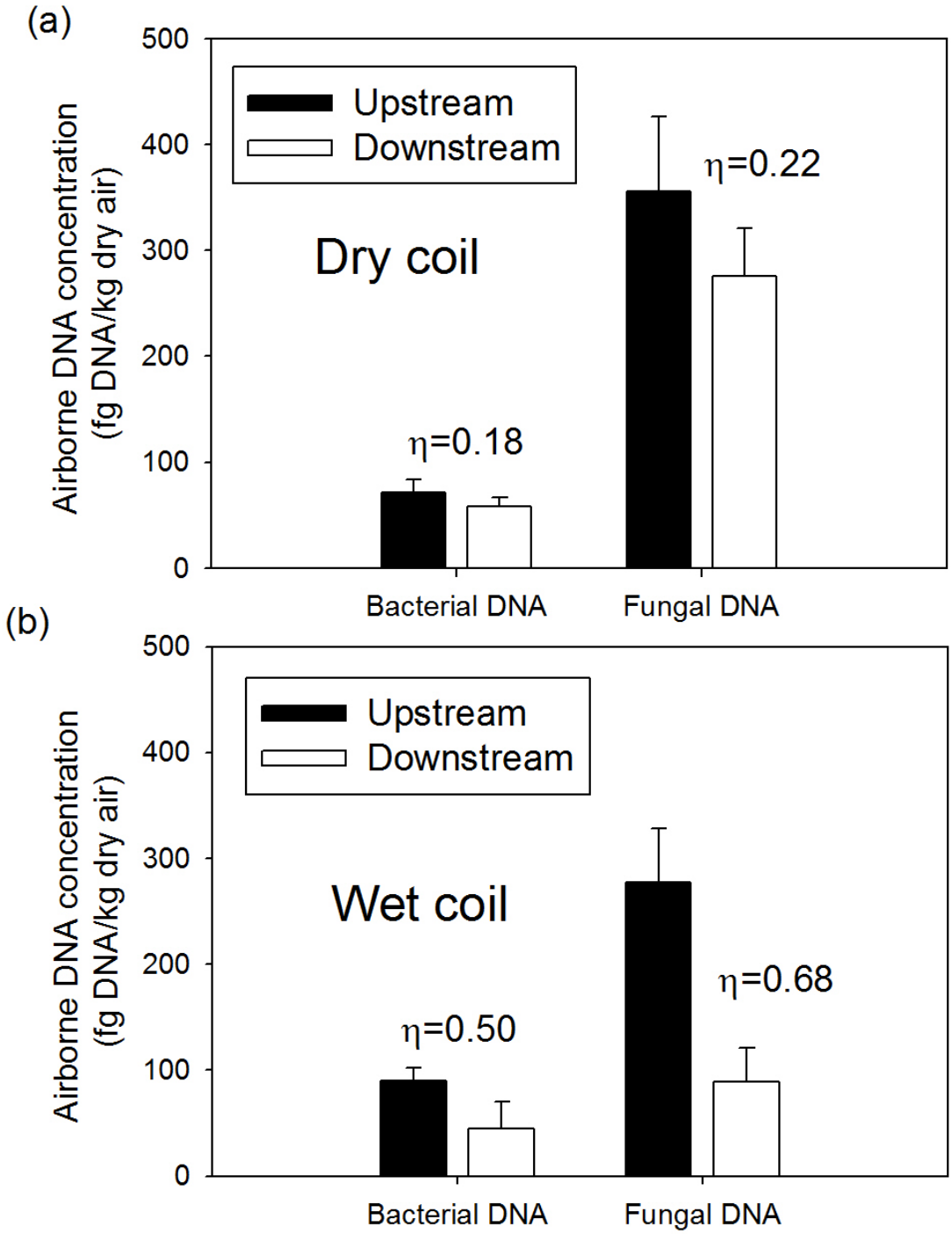
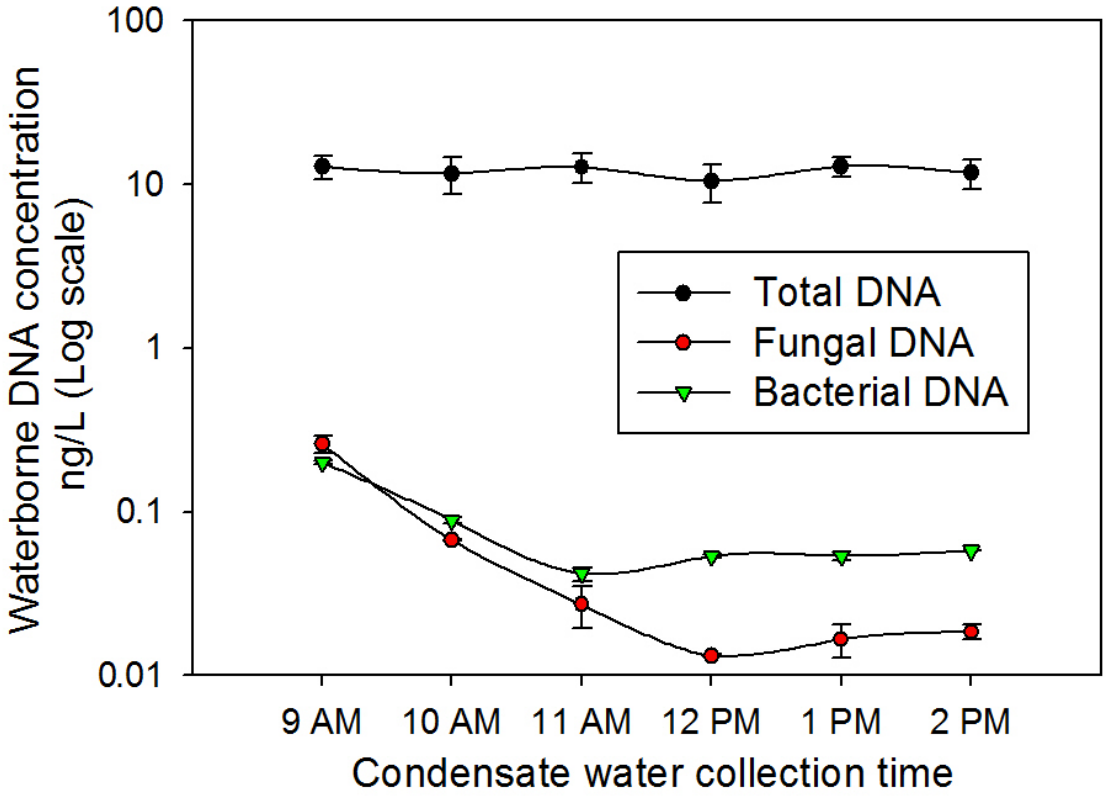


Figure 4. Concentrations of airborne bacterial and fungal DNA sampled upstream and downstream of (a) dry coil and (b) wet coil. The symbol η represents the DNA deposition fraction on the cooling coil. Data points represent averages of nine independent repeated experiments and error bars represent the standard deviations.



632

633 **Figure 5.** Time-series concentrations of total (microbial plus non-microbial), bacterial and
634 fungal DNA in condensate water collected at different daily times with the cooling coil
635 operating for 11 hours per day (from 8 AM to 7 PM). Times are referenced to the collection
636 time of condensate water in one day's operation. Data points represent averages of six
637 independent repeated experiments and error bars represent the standard deviations.
638

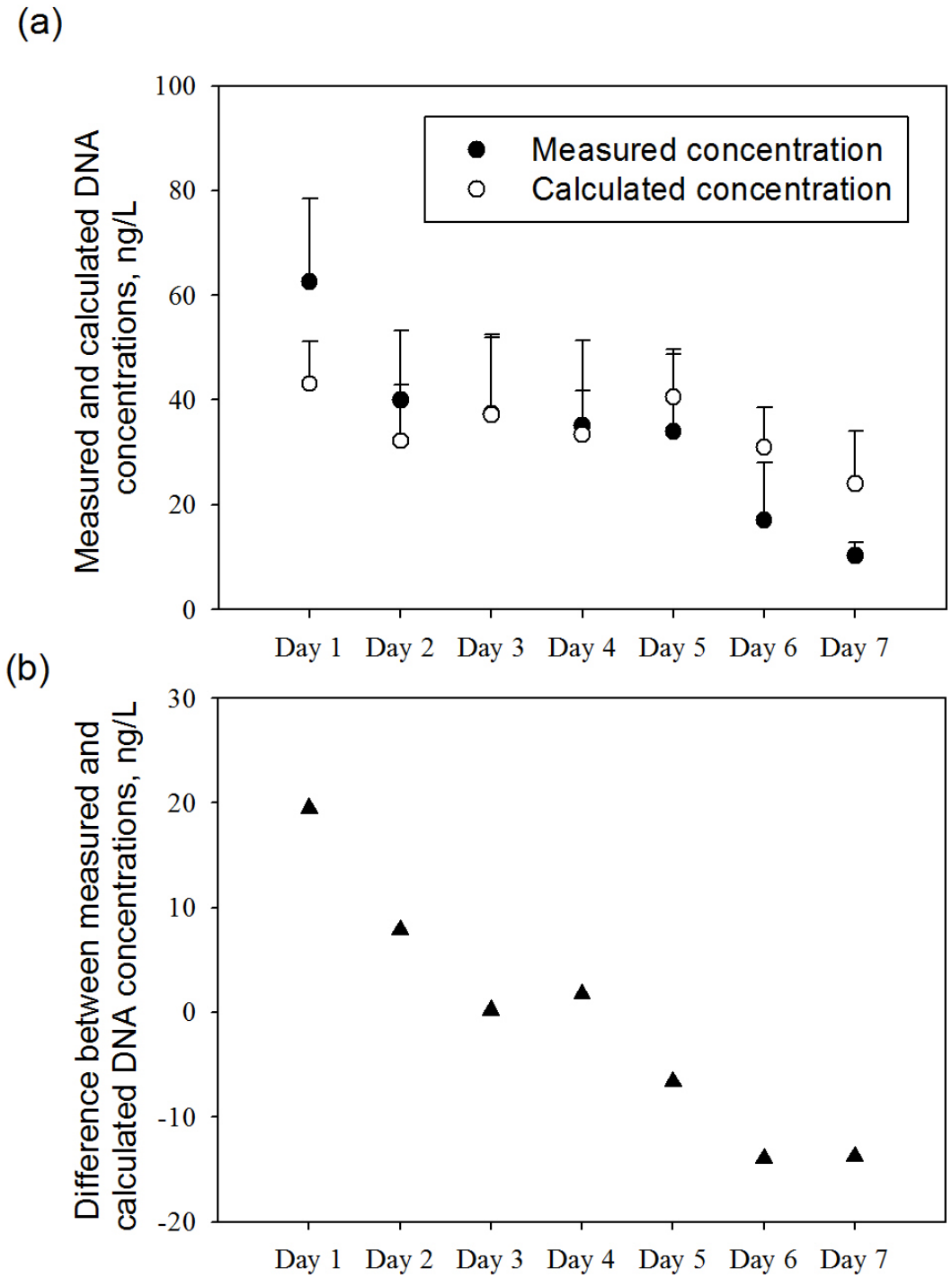


Figure 6. (a) Measured and calculated DNA concentrations in condensate water samples for seven days of continuous operation of the cooling coils. (b) Difference between measured and calculated DNA concentrations in condensate water. Before Day 1, the dry cooling coil had been continually on during daytime for about two weeks. For the seven-day experimental period, the cooling system as well as the fan were operated for 11 hours per day starting at 8 AM, and were off for all other times. For each of seven test days, 3×1 L of condensate water was collected at intervals of 60 min (collection times = 9 AM through 2 PM) and analyzed as described in §2.6. Data points represent averages of three independent repeated experiments and error bars represent the standard deviations for these three measurements.