



# Characterization of naturally occurring airborne diacetyl concentrations associated with the preparation and consumption of unflavored coffee



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## ABSTRACT

Diacetyl, a suspected cause of respiratory disorders in some food and flavorings manufacturing workers, is also a natural component of roasted coffee. We characterized diacetyl exposures that would plausibly occur in a small coffee shop during the preparation and consumption of unflavored coffee. Personal (long- and short-term) and area (long-term) samples were collected while a barista ground whole coffee beans, and brewed and poured coffee into cups. Simultaneously, long-term personal samples were collected as two participants, the customers, drank one cup of coffee each per h. Air sampling and analyses were conducted in accordance with OSHA Method 1012. Diacetyl was detected in all long-term samples. The long-term concentrations for the barista and area samples were similar, and ranged from 0.013–0.016 ppm; long-term concentrations for the customers were slightly lower and ranged from 0.010–0.014 ppm. Short-term concentrations ranged from below the limit of detection (<0.0047 ppm)–0.016 ppm. Mean estimated 8 h time-weighted average (8 h TWA) exposures for the barista ranged from 0.007–0.013 ppm; these values exceed recommended 8 h TWA occupational exposure limits (OELs) for diacetyl and are comparable to long-term personal measurements collected in various food and beverage production facilities. The concentrations measured based on area sampling were comparable to those measured in the breathing zone of the barista, thus exceedances of the recommended OELs may also occur for coffee shop workers who do not personally prepare coffee (e.g., cashier, sanitation/maintenance). These findings suggest that the practicality and scientific basis of the recommended OELs for diacetyl merit further consideration.

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## 1. Introduction

Diacetyl has been used for decades as a flavoring agent to impart a buttery odor and taste in coffee, flour, chocolate, cooking oils, popcorn and other snack foods, dairy products, and baked goods [41,42]. The National Toxicology Program (NTP) has suggested that “human consumption of many foods and beverages containing low levels of diacetyl constitutes a virtually universal exposure scenario for this ubiquitous diketone” [41]. However, concerns have recently been raised regarding apparent increased rates of respiratory disorders in certain food and flavorings manufactur-

ing workers. Specifically, over the past ten years, scientists at the National Institute for Occupational Safety and Health (NIOSH) have investigated numerous microwave popcorn and flavoring production facilities at which diacetyl-containing flavorings were used, and have concluded that diacetyl may be contributing to or causing severe respiratory disorders, including the rare disease *bronchiolitis obliterans*, in highly exposed workers [39]. As a result, diacetyl has largely been phased out of the food flavoring industries and replaced by 2,3-pentanedione and other diketones that also possess “butter-like” qualities [4,5,49].

Due to concerns over worker health and safety, various occupational exposure limits (OELs) for diacetyl have been recommended. In 2011, NIOSH proposed a 15 min Short-Term Exposure Limit (STEL) of 0.025 parts per million (ppm), and an eight hour time-weighted average (8 h TWA) Recommended Exposure Limit (REL)

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of 0.005 ppm for diacetyl [39]. In 2012, ACGIH adopted Threshold Limit Values (TLVs) for diacetyl, 0.02 ppm as a 15 min STEL, and 0.01 ppm as an 8 h TWA [1]. More recently, the European Commission (EC) published draft recommended diacetyl OELs of 0.1 ppm as a 15 min STEL and 0.020 ppm as an 8 h TWA [20]. Similarly, the MAK Commission in Germany has published an 8 h OEL for diacetyl of 0.02 ppm [8]. The NIOSH and the EC recommended OELs have not been finalized to date. Similarly, the U.S. Occupational Safety and Health Administration (OSHA) has not promulgated OELs for diacetyl; however, it was previously listed on the agency's Semi-annual Agenda of Regulations as a long-term action item [47]. In addition, others have previously (i.e., prior to the publication of those listed above) proposed various OELs for diacetyl. Maier et al. used modeling to evaluate the results of studies of diacetyl exposure on respiratory effects in mice to derive an 8 h TWA OEL of 0.2 ppm for diacetyl, which they believe was supported by the current epidemiology literature of diacetyl-exposed workers [31]. Lastly, Egilman et al. proposed an 8 h TWA OEL, based on their evaluation of the available literature, of 0.001 ppm [9].

Diacetyl occurs naturally in many different consumable items, sometimes at concentrations that result in exposures that exceed the aforementioned OELs by several orders of magnitude. For example Pierce et al. (2014) recently reported that naturally occurring diacetyl concentrations in cigarette smoke ranged from 200 ppm to 400 ppm, which results in relatively high exposures even from moderate smoking habits [48]. It was also determined that most of the diacetyl is formed naturally as a byproduct of pyrolysis when tobacco is burned. Similarly, diacetyl is formed when coffee beans are roasted, as a result of amino acids (e.g., glycine and alanine) and sugar molecules (e.g., glucose and mannose) reacting in the bean [7]. This reaction, sometimes referred to as a Maillard reaction, occurs at temperatures of 200 °C or higher, and diketone formation increases with increasing roasting time and temperatures; coffee roasting temperatures typically range from 220 °C to 230 °C [53]. Reported diacetyl concentrations measured in the headspace of stored unflavored roasted coffee beans and ground unflavored roasted coffee have ranged from 0.4 ppm to 4.4 ppm [23,32], and a concentration of 7.0 ppm diacetyl was reportedly measured in the headspace of an open cup of unflavored brewed coffee [57]. While headspace concentrations may not be representative of actual exposures occurring during product use, these studies suggest that coffee baristas and consumers may experience exposures to naturally occurring diacetyl at concentrations in excess of the proposed OELs. This may also hold true for coffee processing workers. Indeed Gaffney et al. (2015), recently found that the mean diacetyl concentration measured during commercial grinding of unflavored coffee exceeded all of the current and proposed short-term OELs, and that the 8 h diacetyl OELs could be exceeded after as little as 7–40 min of grinding [22].

There are thousands of coffee shops in the U.S. at which roasted beans are ground and brewed, and, of course, roasted beans are also ground and brewed in thousands of homes. Accordingly, millions of individuals in the U.S. are potentially exposed to naturally occurring diacetyl on a daily basis, yet to our knowledge, no published studies exist that have evaluated worker or consumer diacetyl exposures during these activities. The purpose of this analysis was to characterize worker and consumer diacetyl exposures that would plausibly occur in a small coffee shop during the preparation and consumption of unflavored coffee. Personal and area samples were collected while a barista ground whole beans, and brewed and poured coffee, and while customers consumed coffee. The airborne data were characterized via comparisons to diacetyl concentrations measured in industrial settings where diacetyl was used as a flavoring agent. We also compared the diacetyl concentrations to the short- and long-term recommended and proposed diacetyl OELs, and discuss the implications of our findings with respect to

the scientific merits of the OELs and claims of disease causation at low exposures. We conclude with suggestions for future areas of research.

## 2. Methods

### 2.1. Coffee used in this study

Two 2.27 kg (5 lb) bags of whole coffee beans were purchased from a local coffee shop. The bags were comprised of aluminized plastic, were sealed, and were stored at room temperature and never refrigerated prior to or during the study. According to the manufacturer, the beans were cultivated in Latin America and were dark roasted at its roastery in California nine days prior to our study. The products' labels described the coffee as caffeinated, medium bodied, unflavored and the retailer's "house blend".

### 2.2. Study location and participants

The study was conducted in duplicate, with one simulation the morning and one in the afternoon, in a 40.3 m<sup>3</sup> (4.4 × 3.5 × 2.6 m) residential kitchen (Fig. 1). The kitchen had 4 doors and 2 operable windows that remained closed during the sampling periods. Two baristas participated in the study, one during each simulation, and four consumers participated in the study, two during each simulation. Between the simulations (approximately 1 h and 50 min) and after the second simulation all of the windows and doors in the room were opened in order to clear the room of any residual diacetyl vapors.

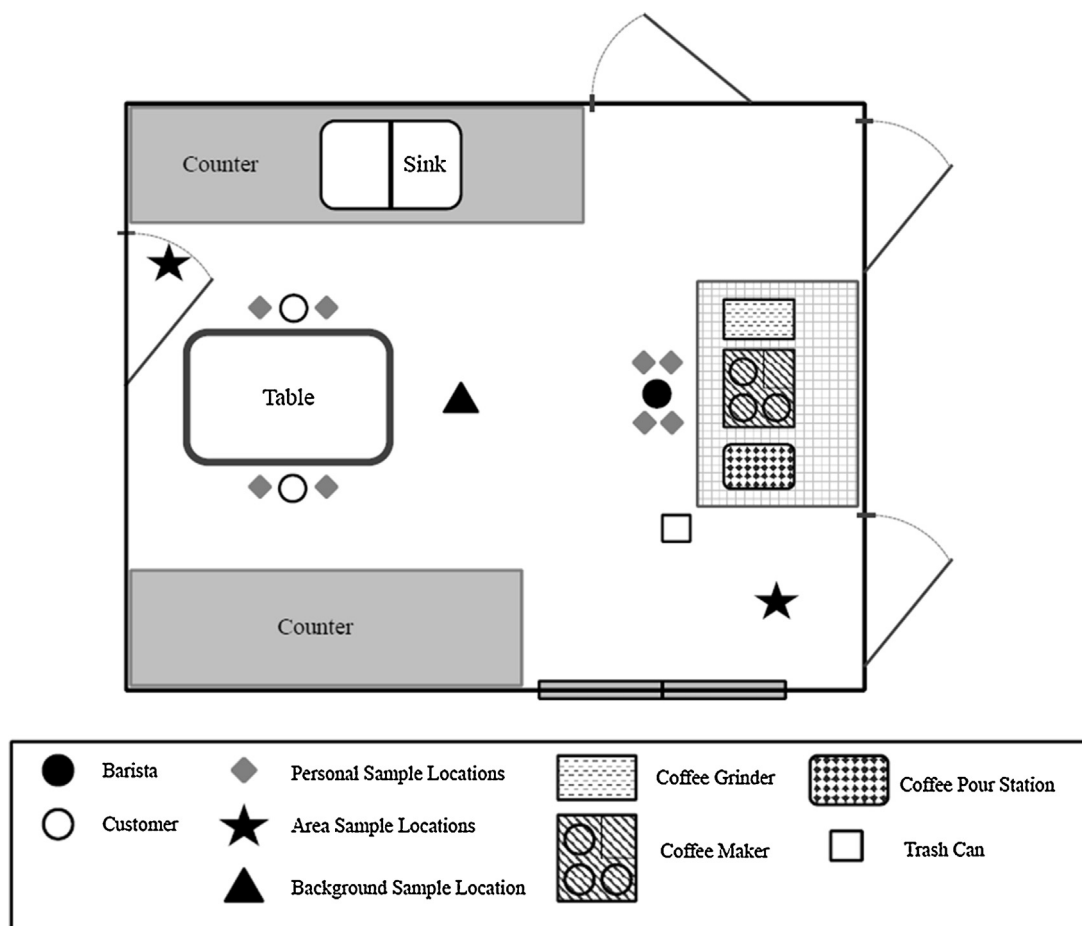
### 2.3. Environmental conditions

Outdoor and indoor temperature and humidity readings were collected prior to each of the simulations using a Q-TRAK Indoor Air Quality Monitor 7565 (TSI, Inc., Shoreview, MN).

### 2.4. Coffee grinding and brewing protocol and barista exposure scenario

At the start of the first simulation, the barista poured the entire 2.27 kg bag of coffee into the hopper of a commercial grinder (Bunn-O-Matic Precision Grinder G9 Series, Springfield, IL), after which the lid to the hopper was closed. During the second simulation the beans remaining in the hopper were visually observed through a clear window in the hopper and were perceived to be running low; therefore, the barista opened the hopper lid, poured an additional 2.27 kg bag of coffee into the hopper, and closed the lid. Approximately 66 g of whole beans were ground per brewed pot of coffee; each grind cycle lasted for less than 10 sec. The ground beans were dispensed by the grinder into an attached metal filter basket containing a paper filter. The coffee grinder was positioned on a counter at a height of 0.91 m.

A commercial coffee brewer (Bunn-O-Matic VP17 Series 3 Burner Coffee Brewer, Springfield, IL) was used by the barista to brew the ground coffee. The coffee brewer was located on the counter within one foot of the grinder, at a height of 0.91 m. Prior to the start of the first simulation, in accordance with the manufacturer's instructions, the brewer's tank was filled to its capacity (approximately 6 L) with water. Immediately after grinding, and prior to each brew cycle, the metal filter basket containing the paper filter and the freshly ground beans, was slid onto the filter basket rails of the brewer. Approximately 1.85 L of water was added to the brewer per cycle, and each brew cycle (time elapsed between addition of water to the brewer until the coffee pot was full) was approximately 4 min. The coffee pot had a filling capacity of 1.85 L of brewed coffee, and the temperature of the brewed coffee was



**Fig. 1.** Schematic of sampling location. Personal and area air samples were collected in several locations throughout the study room in order to characterize diacetyl exposure associated with the preparation and consumption of unflavored coffee.

determined using a TruTemp (Taylor Precision Products, Oakbrook, IL) thermometer to be approximately 82.2 °C. Within 2 min of the end of a brew cycle, the contents of the coffee pot were emptied into 0.23 L (~8 oz) cups (8 cups per pot).

All cups of coffee, except those consumed at the beginning of each hour by the customers, described below, were allowed to sit uncovered on a counter near the brewer for approximately 4 min (to simulate multiple uncovered cups of coffee in a shop) and were then disposed of down a sink drain by the barista; the drain was simultaneously flushed with cold tap water. Prior to each brewing cycle, the barista emptied the used coffee grounds from the previous brew into a 30 L trash can that was equipped with a lid that was opened using a foot pedal, and the trash can was emptied between the two simulations.

Both simulations lasted for 3 h, during which 23 and 24 pots of coffee were brewed, respectively. The frequency with which pots were brewed was consistent with hourly brewing capacity according to the coffee brewer's manufacturer (14.4 L/hr), and was intended to be representative of a small coffee shop.

## 2.5. Customer exposure scenario

Two participants per simulation, designated as the customers, sat across from each other at a small table (height=0.7 m), and remained seated for the duration of each simulation. The customers were located approximately 2 m and 3 m away from the coffee grinder, brewer and garbage can containing the disposed grounds. The customers ingested 0.23 L (1 cup) of coffee each per hour.

## 2.6. Diacetyl sampling protocol and analytical methods

### 2.6.1. Short-term sample collection

Prior to the study, a 30 min background sample was collected at approximately breathing zone height (1.4 m) in the middle of the room (Fig. 1). Two short-term (~15 min) personal samples were collected in the breathing zone of the barista at the beginning of each hour during both simulations (12 samples total). The samples were collected on the right and left lapels (6 samples total on each side) of the barista, during which time the barista ground whole beans, brewed two pots and poured 16 cups of coffee.

### 2.6.2. Long-term sample collection

During each simulation, two long-term (3 h) personal samples were collected in the breathing zone of the barista (4 samples total). Similarly, during each simulation, two long-term (3 h) personal samples were collected in the breathing zone of each customer (8 samples total). Finally, four long-term (3 h) area samples were collected at approximately breathing zone height (1.4 m) at locations 1 m and 3.2 m distant from the barista's work area (to the right of the work area and behind the work area). One sample was collected in each location during each simulation. The area samples were intended to represent potential occupational bystander exposures during coffee preparation (e.g., other coffee shop employees). A schematic of these locations is presented in Fig. 1.

### 2.6.3. Analytical methods

The air sampling and subsequent laboratory analysis were conducted in accordance with OSHA Method 1012 [45]. All air samples

for diacetyl were collected on two silica gel sorbent tubes connected in series (SKC Inc., Eighty Four, PA) using GilAir Plus air sampling pumps (Gilian, St. Petersburg, FL). Long- and short-term samples were collected at airflow rates of approximately 0.05 liters per minute (LPM) and 0.2 LPM, respectively. Blank silica gel tubes were provided to the laboratory for quality control purposes; diacetyl-spiked tubes were not provided to the laboratory for analysis. The sampling pumps were calibrated with a Bios DryCal DC-Lite primary flow calibrator (Bios International Corporation, Butler, NJ) before and after sample collection.

During and after sampling, in accordance with the sampling method, all samples were shielded from ultra-violet light using aluminum foil, to avoid the decomposition of the diacetyl. All sampling media were sealed immediately after each sampling event, packaged with freezer ice packs, and sent to an American Industrial Hygiene Association (AIHA) accredited laboratory (ALS Global, Salt Lake City, UT). The limit of quantification was 0.050 µg per sample, corresponding to limits of detection (LODs) for the air samples varying from 0.0013 ppm to 0.0051 ppm.

### 2.7. Statistical analysis

A value corresponding to one half the LOD was used as a substitute for samples found to be less than the LOD for the calculation of arithmetic means. Paired t-tests were used to compare the mean concentrations for short-term samples collected in the first, second and third hours of both simulations. One-way ANOVA was used to compare the long-term concentrations for the barista, customer and area samples.

### 2.8. Estimation of eight-hour time-weighted average diacetyl concentrations

#### 2.8.1. Coffee preparation

We estimated the corresponding 8 h TWA concentrations for the barista using two exposure scenarios.

In the first exposure scenario (“full work shift”), we assumed that the barista left the work area for one hour (i.e., took two 15 min breaks and a 30 min lunch break) over the course of the 8 h work shift. During this time, exposure was assumed to be zero. For the remaining 7 h of the work shift, we assumed the exposure was equivalent to the concentrations (min, max and mean) measured based on the long-term (3 h) samples collected in the breathing zone of the barista.

For the second scenario (“half work shift”), we assumed that the barista prepared coffee for 4 h, during which the exposure was equivalent to the concentrations (min, max and mean) measured based on the long-term (3 h) samples collected in the breathing zone of the barista. We assumed that no additional diacetyl exposure occurred during the remaining 4 h of the work shift.

#### 2.8.2. Coffee shop customers

Because coffee shop customers may spend a few minutes to a few hours on the premises, we evaluated several exposure durations ranging from 3 min to 3 h. We believe this range likely captures the visit duration for the vast majority of those who frequent a coffee shop. For each exposure duration we assumed the exposure was equivalent to the concentrations (min, max and mean) measured based on the personal long-term (3 h) customer samples and zero for the remainder of the 8 h work day (i.e., the ‘remainder of the work day’ represents time *not* spent in a coffee shop [range: 300–477 min]).

### 2.9. Evaluation of representative work shift diacetyl concentrations in facilities using diacetyl-containing flavorings

Numerous studies of workday-duration exposure to diacetyl have been conducted in a variety of facilities, including dairy and commercial coffee processing, and microwave popcorn and snack food manufacturing facilities, where diacetyl-containing flavorings were used, oftentimes in high volumes [10–19,35–37,39,40,56]. The data for the studies conducted by the Eastern Research Group (ERG) are available from the U.S. federal government [46]. For the purposes of this analysis, we extracted the results of individual or composite (multiple samples collected consecutively on one worker over the course of a work shift) personal samples collected for a total of at least 6 h in these facilities. Samples analyzed using NIOSH Method 2557 were excluded, as the results of such analyses have been found to be significantly influenced by temperature, humidity and time to extraction [6]. However, some historical data have since been “corrected” for these parameters, using the method described by Cox-Ganser et al. [6]; such data were reported by NIOSH and were included in our analysis [39]. Descriptive statistics, including the unweighted mean, median, and various percentiles of the samples collected on workers who likely personally handled diacetyl-containing flavorings were tabulated. When possible, the sampling results from individual samples were extracted and used in this analysis. However, certain studies only reported the range and the mean of the sampling results. In these instances, only the mean concentration was used in our statistical analysis.

## 3. Results

A total of 47 pots of coffee were brewed, and 376 cups of coffee were poured during the two simulations. A total of 29 samples were collected and analyzed for diacetyl, consisting of 12 short-term personal, 12 long-term personal, 4 long-term area, and 1 background sample (Table 1).

### 3.1. Environmental conditions

The outdoor temperature and relative humidity prior to the first simulation were 5.28 °C and 62.5%, respectively. Indoor, the temperature and relative humidity prior to the first simulation were 19.6 °C and 46%. The outdoor temperature and relative humidity prior to the second simulation were 11.3 °C and 40.9%, respectively. Indoor, the temperature and relative humidity prior to the second simulation were 19.6 °C and 43.6%.

### 3.2. Diacetyl sampling results

The background diacetyl concentration in the study area prior to the first simulation was less than the LOD (<0.0024 ppm). There were no statistically significant differences in the sampling results for the first versus the second simulation, nor were there any differences in right versus left lapel measurements collected on the barista and on the customers. Therefore, unless otherwise noted, the results were combined.

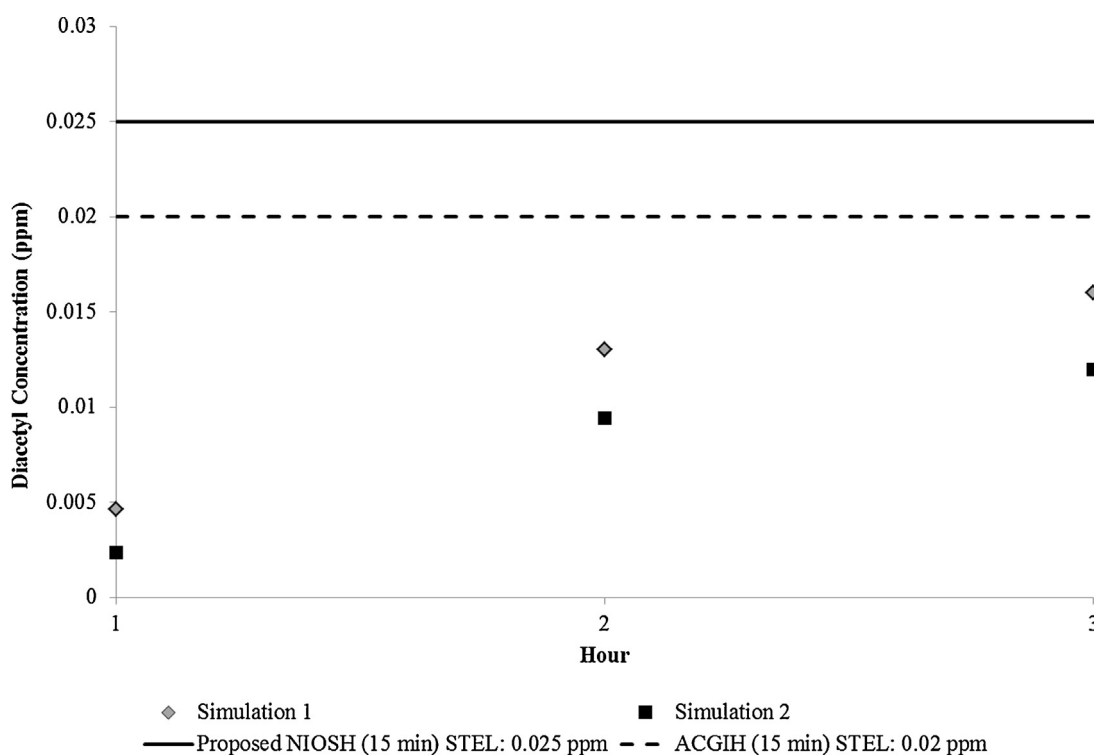
#### 3.2.1. Short-term diacetyl concentrations

Diacetyl concentrations for the personal short-term samples ranged from below the LOD (<0.0047 ppm)–0.016 ppm, with an overall mean of 0.010 ppm (Table 1). Diacetyl was detected in 9 samples; 3 samples were non-detect, all of which were collected during the first hour of both simulations. It is interesting to note that the mean short-term diacetyl concentrations increased significantly throughout the course of each simulation (Fig. 2). Specifically, the mean short-term concentration measured in the third hour of the simulations was statistically significantly higher

**Table 1**  
Diacetyl concentrations measured during the preparation and consumption of coffee. Personal and area air samples were collected to characterize potential exposures to coffee shop workers and customers associated with the preparation and consumption of unflavored coffee, respectively. Statistical comparisons were performed using paired *t*-tests. Diacetyl was detected in all long-term samples, and there were no statistically significant differences in the mean long-term diacetyl concentrations measured for the barista versus the customer versus the area samples. Diacetyl was detected in three-quarters of the short-term samples, and the short-term concentrations increased significantly throughout the course of the study.

| Sample description | Number of samples | Sample duration (min) | Concentration (ppm) |         |                    |                    |
|--------------------|-------------------|-----------------------|---------------------|---------|--------------------|--------------------|
|                    |                   |                       | Minimum             | Maximum | Mean               | Standard deviation |
| Background         | 1                 | 30                    |                     |         | <0.0024            |                    |
| <i>Long Term</i>   |                   |                       |                     |         |                    |                    |
| Barista            | 4                 | 180                   | 0.013               | 0.016   | 0.015              | 0.0017             |
| Customer           | 8                 | 180                   | 0.010               | 0.014   | 0.013              | 0.0017             |
| Area               | 4                 | 180                   | 0.013               | 0.016   | 0.015              | 0.0015             |
| <i>Short Term</i>  |                   |                       |                     |         |                    |                    |
| Barista            |                   |                       |                     |         |                    |                    |
| 1st hour           | 4                 | 15                    | <0.0047             | 0.007   | 0.003 <sup>a</sup> | 0.0023             |
| 2nd hour           | 4                 | 15                    | 0.009               | 0.014   | 0.011              | 0.0022             |
| 3rd hour           | 4                 | 15–16                 | 0.012               | 0.016   | 0.014              | 0.0023             |
| Overall            | 12                | 15–16                 | <0.0047             | 0.016   | 0.010              | 0.0051             |

<sup>a</sup> For samples below the LOD, 1/2 the LOD was used when calculating descriptive statistics.



**Fig. 2.** Short-term diacetyl concentrations measured during coffee preparation. Short-term air samples were collected in the breathing zone of the barista at the start of each hour over both simulations. Diacetyl concentrations ranged from below the LOD (<0.0047 ppm)–0.016 ppm, and mean short-term diacetyl concentrations increased significantly throughout the course of each simulation.

than the mean of the second hour ( $p=0.0071$ ) and first hour ( $p=0.0021$ ), and the mean short-term concentration measured in the second hour was statistically significantly higher than the mean of the first hour measurements ( $p=0.0013$ ).

### 3.2.2. Long-term diacetyl concentrations

The long-term (3 h) diacetyl concentrations were relatively consistent and far less variable than the short-term concentrations (Table 1). Diacetyl was detected in all of the long-term personal barista samples; concentrations ranged from 0.013 ppm to 0.016 ppm, with a mean of 0.015 ppm. Diacetyl was also detected in all of the long-term personal customer samples; concentrations ranged from 0.010 ppm to 0.014 ppm, with a mean of 0.013 ppm.

Diacetyl was also detected in all of the long-term area samples; concentrations ranged from 0.013 ppm to 0.016 ppm, with a mean of 0.015 ppm (Table 1). There was no apparent difference between the mean concentrations measured in the area samples collected at the location closer to the barista's work area (0.015 ppm at 1 m) versus the more distant location (0.014 ppm at 3.2 m). Overall, there were no statistically significant differences in the mean long-term diacetyl concentrations measured for the barista versus the customer versus the area samples ( $p=0.14$ ).

### 3.2.3. Comparisons of sampling results to proposed or recommended diacetyl OELs

None of the individual short-term (15 min) barista samples (maximum of 0.016 ppm) exceeded the proposed NIOSH or ACGIH

**Table 2**

Estimated 8 h TWA diacetyl concentrations associated with the preparation and consumption of coffee. To compare the measured diacetyl concentrations to the proposed or adopted OELs, 8 h TWA diacetyl exposures were estimated for the barista and customers assuming various plausible durations of coffee preparation or consumption. The estimated 8 h TWA concentrations for the barista, assuming that they worked a full or half work shift exceeded the proposed NIOSH 8 h REL, and the ACGIH 8 h TLV was exceeded for the full work shift scenario. Exposures to coffee shop customers could exceed the draft NIOSH 8 h REL of 0.005 ppm after approximately 3 h in the shop.

| Scenario | Duration of coffee preparation or consumption per day | Estimated 8 h TWA (ppm) |         |         |
|----------|---|-------------------------|---------|---------|
|          |   | Minimum                 | Maximum | Mean    |
| Barista  | 7 h (full work shift)                                 | 0.011                   | 0.014   | 0.013   |
|          | 4 h (half work shift)                                 | 0.007                   | 0.008   | 0.007   |
| Customer | 3 min   | 0.00006                 | 0.00009 | 0.00008 |
|          | 30 min  | 0.0006                  | 0.0009  | 0.0008  |
|          | 3 h   | 0.004                   | 0.005   | 0.005   |

STELs (0.025 ppm and 0.02 ppm, respectively). All individual long-term (3 h) barista, customer and area measurements were at least two-fold higher than the proposed NIOSH 8 h REL (0.005 ppm) and met or exceeded the ACGIH 8 h TLV (0.01 ppm) for diacetyl. None of the long-term measurements exceeded the diacetyl 8 h OEL recommended by the EC and the MAK Commission (0.02 ppm).

As shown in Table 2, the estimated 8 h TWA concentrations for the barista ranged from a mean of 0.007 ppm for the half work shift to a mean of 0.013 ppm for the full work shift. These estimated 8 h TWAs exceeded the proposed NIOSH 8 h REL, and the ACGIH 8 h TLV was exceeded for the full work shift scenario. According to the data collected in this analysis, coffee shop customers could exceed the draft NIOSH 8 h REL of 0.005 ppm after approximately 3 h in the shop (Table 2).

### 3.2.4. Comparison of barista 8 h TWA diacetyl concentrations to long-term concentrations in food production facilities

The results of all long-term individual and composite personal samples (the total sampling duration was  $\geq 6$  h) collected in food production facilities in which diacetyl or diacetyl-containing flavorings were handled are summarized in Appendix A. A total of 98 data points, which represented 838 long-term exposure measurements were collected in 15 establishments: four microwave popcorn facilities, two bakeries, two facilities producing baked snack foods, five facilities producing dairy products, and one coffee processing plant. Samples were collected on a variety of workers, including those involved in mixing, packaging, warehouse operations, maintenance, quality assurance, sanitation, as well as supervisors.

A total of 175 long-term measurements were reported for workers who likely directly handled diacetyl or diacetyl-containing flavorings (e.g., mixers, cooks, flavoring workers, blender operators) (Fig. 3). The long-term diacetyl concentrations ranged from 0.00063 ppm to 2.70 ppm (the concentrations for 3 measurements were reported as <0.002 ppm, which may be lower than 0.00063 ppm), with a mean of 0.30 ppm (standard deviation = 0.60 ppm). The median concentration was 0.011 ppm, whereas the 25th and 75th percentile concentrations were 0.003 ppm and 0.12 ppm, respectively. The mean estimated 8 h TWA concentrations of diacetyl for a barista (0.007 ppm and 0.013 ppm) correspond to the 42nd and 53rd percentiles of this dataset.

In summary, our data indicate that barista exposures to naturally occurring diacetyl may be comparable to those experienced in settings where diacetyl-containing flavorings are handled and used for food and beverage production.

## 4. Discussion

Unroasted green coffee beans contain little to no diacetyl [7]. When coffee beans are roasted to achieve the desired aroma and flavor profile of the brewed coffee, volatile diacetyl and other dike-

tones (e.g., 2,3-pentanedione) are formed inside the beans. The grinding of roasted coffee beans then releases the diacetyl in vapor form. The findings of earlier investigations (e.g., [22,57]) suggested that exposure to naturally occurring diacetyl might occur in retail establishments where coffee is roasted and/or freshly ground. In this study, we simulated a small coffee shop setting in which a barista ground, brewed, and poured numerous cups of unflavored coffee over a period of 3 h. To the best of our knowledge, this is the first published study to describe airborne concentrations of naturally formed diacetyl associated with the preparation and consumption of unflavored coffee.

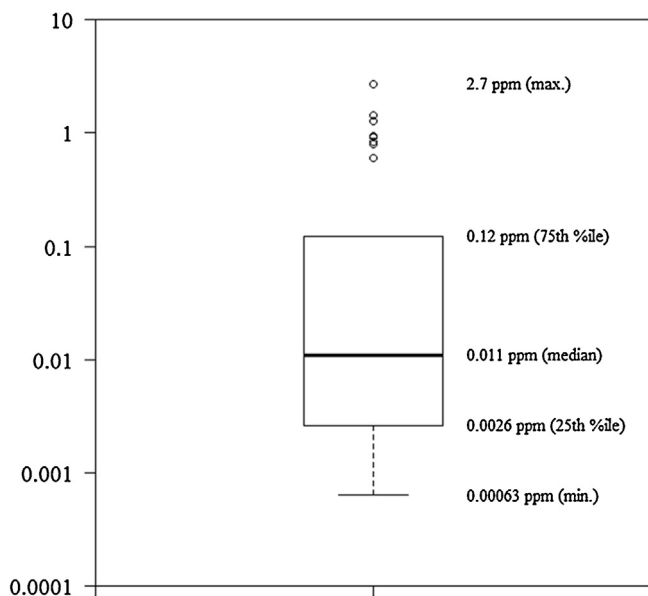
### 4.1. Summary of our findings

Diacetyl was readily detectable in the short-term (15 min) and long-term (3 h) personal samples collected on the barista (Table 1). Interestingly, the short-term barista concentrations increased over time (Fig. 2) and were statistically significantly higher at the third hour than in either the second or first hours. The long-term diacetyl concentrations collected for the duration of each simulation were similar regardless of location. Specifically, all long-term personal barista and customer samples, and all of the area samples from the two different locations fell within a very narrow range of 0.010 ppm to 0.016 ppm (Table 1).

Overall, these findings suggest that diacetyl emitted from the various sources, particularly those near the barista (i.e., associated with grinding, brewing, uncovered cups of coffee) quickly mixed throughout the room, such that the near field and far field concentrations of diacetyl were similar. Based on the trend of increasing short-term concentrations over time, it is evident that airborne diacetyl accumulated in the room over the course of the simulations, which is likely due to a low air exchange rate in the study room. This is consistent with the fact that all doors and windows were kept closed throughout the study. If indeed the airborne diacetyl levels in the study area had not equilibrated to steady state conditions by the end of the study period, then it seems plausible that the diacetyl concentration in the room would have continued to increase if the simulation study had continued beyond 3 h. Interestingly, the findings also suggest that diacetyl accumulation in the room was a larger contributor to the short-term measurements than coffee preparation (e.g., grinding, brewing and pouring activities) directly.

### 4.2. Comparison to diacetyl measurements in other studies

Gaffney et al. measured 0.018–0.39 ppm of naturally occurring diacetyl in short-term (8–11 min) area samples collected at breathing zone height next to a commercial grinder that processed 11 kg of freshly roasted, unflavored coffee beans during the sampling event [this corresponds to about 180 L (i.e., 300–400 medium-sized cups) of brewed coffee] [22]. The short-term personal diacetyl concentrations measured on the barista in this study were lower than the



**Fig. 3.** Long-term ( $\geq 6$  h) personal measurements collected in food production facilities in which diacetyl or diacetyl-containing flavorings were handled. The results of long-term ( $\geq 6$  h duration) personal samples collected on workers who likely directly handled diacetyl or diacetyl-containing flavorings in food and beverage production facilities were extracted from published literature and unpublished reports. The estimated 8 h TWA concentrations of diacetyl for a barista (0.007 ppm and 0.013 ppm) correspond to the 42nd and 53rd percentiles of this dataset.

values reported by Gaffney et al. [22], and this is certainly due in part to the much smaller volumes of coffee processed (approximately 1.4–1.5 kg total in 3 h) in the current study. In addition, the coffee beans used in the Gaffney et al. study had been roasted on the premises only a few hours before grinding, while in the current study the beans were roasted 9 days prior to the study and had been shipped from a distant location; therefore, some diacetyl loss may have occurred before the beans were ground. Finally, Gaffney et al. reported that airborne diacetyl concentrations decreased significantly with distance from the grinder; that trend was not observed in the current study and this is likely due to the fact that the Gaffney et al. study took place in a large commercial facility, while the current study was conducted in a much smaller room with relatively little air exchange.

The mean estimated 8 h TWA diacetyl concentrations for the barista (0.007 ppm and 0.013 ppm for the half- and full-shift estimates, respectively) were comparable to concentrations based on long-term ( $\geq 6$  h duration) personal samples collected in various food and beverage production facilities (Fig. 3). The values that comprise the distribution illustrated in Fig. 3 were collected specifically to evaluate personal diacetyl 8 h TWA exposures in occupational settings where diacetyl-containing flavorings were known to be used, and the vast majority of these surveys were conducted in response to the initial reports of respiratory disease in workers at a microwave popcorn packaging plant where diacetyl was present in artificial butter flavoring (e.g., [28]). Many of these surveys involved collection of industrial hygiene data only, i.e., worker health was not evaluated. In short, daily 8 h TWA diacetyl exposures experienced by a barista in a coffee shop may not be much different from those that occur in some facilities where diacetyl is handled. This is likely due in part to the fact that (1) most diacetyl-containing flavorings are actually fairly dilute (e.g., 1–3% diacetyl), and (2) the diacetyl emitted from ground beans is in a vapor phase, while the flavorings are powders, solids, and liquids at room temperature.

#### 4.3. Comparison to proposed diacetyl OELs: potential implications for coffee shop workers and customers

Mean long-term (3 h) diacetyl measurements for the personal barista and customer samples, as well as the area samples, ranged from 0.013 ppm to 0.015 ppm. Accordingly, under the conditions employed in this study, the proposed NIOSH 8 h TWA REL of 0.005 ppm would be exceeded (regardless of location in the room) after approximately 3 h even if no diacetyl exposure occurred for the remainder of the day. While few customers are likely to remain in a coffee shop for 3 h, certainly most coffee shop employees would be present for at least this amount of time. Indeed, the estimated 8 h TWA exposure for a barista, based on the results of our study in which coffee was prepared for a total of 3 h in one day, met (simulation 2) or exceeded (simulation 1) the proposed NIOSH 8 h TWA REL. Further, if diacetyl levels did continue to increase throughout a large portion of the day, then it is possible that the ACGIH 8 h TWA OEL of 0.010 ppm, and perhaps even recommended STELs, could be exceeded by employees or customers. Interestingly, if the results of our study are representative of a typical coffee shop, then the 8 h TWA OEL of 0.001 ppm proposed by Egilman et al. would likely be exceeded even by customers who are present in the shop for a relatively short amount of time (again, assuming no diacetyl exposure for the remainder of the day) [9].

Of course, these observations are applicable primarily to the conditions of our study; measurements collected in different settings (with varying room dimensions, air exchange rates, volume of coffee processed, etc.) would be required to better characterize potential diacetyl exposures, and to evaluate whether OEL exceedances would occur in operating coffee shops. Nonetheless, it is worthwhile to consider the potential implications if indeed the proposed NIOSH or ACGIH diacetyl OELs were to be adopted by OSHA, given that the enforceable standard might be routinely exceeded in coffee shops. We are unaware of any other instances in which naturally occurring compounds in food products exceed enforceable occupational standards in retail/consumer settings. While the implementation of engineering and personal protective measures to reduce exposures below OELs may be feasible in commercial operations where industrial hygiene principles (e.g., protocols for chemical handling, etc.) are understood and consistently monitored and implemented, it is highly unlikely any such measures could be practically installed and enforced in the thousands of smaller scale operations (e.g., local coffee shops, roasting/grinding operations in grocery stores, etc.) that currently operate in the U.S.

#### 4.4. Does naturally occurring diacetyl in consumer products pose a risk of serious lung disease?: Scientific underpinnings of the proposed diacetyl OELs

The NIOSH and ACGIH diacetyl OELs were derived from exposure-response analyses of respiratory obstruction in a cohort of workers at the Gilster-Mary Lee (GML) microwave popcorn plant in Jasper, Missouri, where diacetyl was present in artificial butter flavoring. These OELs proposed by NIOSH and ACGIH were based on protection against lung damage, respiratory obstruction and *bronchiolitis obliterans*; NIOSH estimates that the risk of pulmonary function decline associated with their OELs would be less than 1 in 1000 in workers ([39], p. 129). As described below, however, it seems clear that these proposed OELs may be routinely exceeded as a result of naturally occurring sources, but with no attendant risk of disease.

For example, coffee processing workers can be exposed to natural diacetyl levels well above the OELs [22], yet the respiratory status of coffee processing workers has been evaluated in many studies [21,26,27,29,33,43,50–52,54,55,58,59] and

the most consistent adverse effects are allergic respiratory responses to specific allergens in respirable green coffee dusts [21,26,27,29,30,50,54,55,59]. To the best of our knowledge, there are no reports of *bronchiolitis obliterans* or other serious, obstructive diseases occurring at elevated rates in coffee processing workers handling unflavored coffee beans or ground coffee. Similarly, although retail coffee workers may be exposed to naturally occurring diacetyl at levels exceeding the recommended OELs (as suggested by the present analysis), we are not aware of any epidemiology studies suggesting an increased risk of obstructive disorders in coffee shop workers, nor are there any published case reports of *bronchiolitis obliterans* occurring in coffee shop employees. Finally, cigarette smoke contains naturally occurring diacetyl concentrations in the hundreds of parts per million, and the resulting exposures to smokers are much higher than those typically experienced by workers handling diacetyl-containing flavorings in food processing facilities. However, smoking has not been associated with an increased risk of *bronchiolitis obliterans* [48].

We acknowledge that OELs derived by NIOSH and ACGIH are based on toxicological risk assessment methodologies that do not involve considerations of practicality or feasibility. Nonetheless, we believe it is worthwhile to examine the underlying analyses in this case in order to appreciate how it is possible to derive “health-based” OELs that are actually below naturally occurring levels associated with handling a commonly used food product (coffee). First, the reported findings of a diacetyl-related increased incidence of serious respiratory disease in the GML cohort should be reevaluated. Specifically, while Kreiss et al. [28] reported an increased prevalence of respiratory obstruction, subsequent studies of the GML cohort failed to confirm any increased risk of this endpoint [2,3]. Even if one assumes that the GML cohort had an increased risk of obstruction, as has been noted by others it is unclear how these effects could all be ascribed to workplace diacetyl exposure, because: (1) there were hundreds of volatile organic compounds measured at the facility, many of which were known respiratory irritants [31,38], (2) a significant fraction (57%) of the workforce indicated they were exposed to respiratory irritants at off-site locations, including chemicals known to be causes of *bronchiolitis obliterans* (e.g., nitrogen oxides found in silo gas) [28], and (3) rates of respiratory obstruction were actually more highly correlated with respirable dust exposure than diacetyl exposure [38]. As noted earlier, OSHA has not promulgated a standard to govern occupational exposure to diacetyl, and they have concluded that “A cause-effect relationship between diacetyl and bronchiolitis obliterans is difficult to assess because . . . food-processing and flavor-manufacturing employees with this lung disease were exposed to other volatile agents” [44].

#### 4.5. Study strengths and limitations and areas of future research

The primary limitation of this study was that only one location was evaluated, and our results are likely to be most relevant to other settings with similar characteristics. For example, room dimensions, air exchange rates, volume of coffee processed, and other factors are likely to influence airborne diacetyl concentrations, and these parameters are certain to vary significantly among other settings. Hence, the direct applicability of our findings to “real world” settings (e.g., retail coffee shops) is unclear and therefore our study should be considered a pilot or “hypothesis generating” effort.

The major strength of the current study is that it addresses a significant information gap regarding the feasibility of implementing proposed diacetyl OELs. Also, the variability in the measured concentrations over the course of the duplicate sampling runs was minimal. Future research efforts should focus on additional measurements of naturally occurring diacetyl in other (simulated or actual) retail settings where coffee is roasted and/or ground

for consumers. It would also be helpful to assess the degree to which diacetyl exposures associated with coffee preparation might occur in the home. Lastly, alternative approaches to establishing diacetyl OELs should be considered. Specifically, health effect surveys conducted in workplaces where diacetyl-containing flavorings are used are known to be confounded by co-exposures to many other agents, and therefore analyses of exposure-response relationships based on animal diacetyl inhalation studies [24,25,34], such as the approach recently proposed by Maier et al. [31], would likely provide a more robust basis for a practical, yet health-protective OEL.

## 5. Conclusions

Our results indicate that coffee shop workers and customers are exposed to naturally occurring diacetyl during the preparation and consumption of unflavored coffee. All long-term measurements and estimated 8 h TWA exposures met or exceeded proposed and adopted OELs set forth by NIOSH and ACGIH. Before any significant investment is made to evaluate cost-effective measures to protect coffee workers from exposures to naturally occurring diacetyl, it would seem prudent to assess the scientific merits of the OELs themselves, as well as any underlying assertions that a causative relationship between diacetyl exposure and serious respiratory disorders (including *bronchiolitis obliterans*) has been observed.

## Acknowledgements and Conflicts of Interest

All the authors are employed by Cardno ChemRisk, a consulting firm that provides scientific advice to the government, corporations, law firms and various scientific/professional organizations. Cardno ChemRisk has been engaged by several manufacturers and suppliers of diacetyl and diacetyl-containing flavorings in various litigation matters, and two of the authors (Drs. Pierce and Finley) have served as experts in diacetyl litigation. However, no external funding was received for the study, the research supporting the analysis, nor the time needed to prepare the article.

## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.toxrep.2015.08.006>.

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