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An Exploratory Study: Assessment of Modeled Dioxin Exposure in Ceramic Art Studios

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National Center for Environmental Assessment
Office of Research and Development
U.S. Environmental Protection Agency
Washington, DC

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ABSTRACT

The purpose of this report is to describe an exploratory investigation of potential dioxin exposures to artists/hobbyists who use ball clay to make pottery and related products. Dermal, inhalation and ingestion exposures to clay were measured at the ceramics art department of Ohio State University in Columbus, OH. The measurements were made in two separate studies, one in April 2003 and one in July 2004. This assessment combines the results of these two studies. Estimates of exposure were made based on measured levels of clay in the studio air, deposited on media representing food and on the skin of artists. Dioxin levels in the clay were based on levels reported in the literature for commercial ball clays commonly used by ceramic artists.

Hypothetical dioxin dose estimates were calculated for each subject assuming that all used a 20% ball clay blend with 162 pg TEQ/g. The single-day total doses across the 10 subjects were estimated to range from 0.49 to 20.81 pg TEQ/day, with an average of 3.45 pg TEQ/day. The dermal dose was the major contributor to total dose, exceeding 78% for all subjects. A Monte Carlo simulation suggested that ball clay exposures in a broad population of artists could extend to levels lower or higher than the levels estimated for the 10 subjects. Comparing US average background intakes (adjusted to an absorbed basis) to the 10 subject average dose from ball clay use, indicates that the average ball clay dose is 10% of the background CDD/CDF dose (34.4 pg TEQ/day).

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LIST OF ABBREVIATIONS AND ACRONYMS

˚C	degrees Centigrade
CDD	Chlorinated dibenzo-*p*-dioxin
CDD/F	Chlorinated dibenzo-*p*-dioxins and chlorinated dibenzofurans
CDF	Chlorinated dibenzofuran
cm	centimeter
d	day
DI	Deionized
EDS	Energy dispersive spectroscopy
EPA	U.S. Environmental Protection Agency
ET	Extrathoracic
FDA	U.S. Food and Drug Administration
g	gram
GFF	Glass fiber filters
HpCDD	Heptachlorodibenzo-*p*-dioxin
hr	hour
HRMS	High-resolution mass spectrometry
HxCDD	Hexachlorodibenzo-*p*-dioxin
IRB	Institutional Review Board
kg	kilogram
Kow	Octanol-water partition coefficient
L	liter
L/min	liters per minute
LRB	Laboratory record book
m	meter
mg	milligram
min	minute
mL	milliliter
mm	millimeter
MMAD	Mass median aerodynamic diameter
ND	Nondetect
ng	nanogram
NR	Not reported

LIST OF ABBREVIATIONS AND ACRONYMS (continued)

OCDD	Octachlorodibenzo-*p*-dioxin
OSHA	Occupational Safety and Health Administration
OSU	Ohio State University
oz	ounces
PCDD	Polychlorinated dibenzo-*p*-dioxin
PCDF	Polychlorinated dibenzofuran
PCD/F	Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans
PeCDD	Pentachlorodibenzo-*p*-dioxin
pg	picogram
PU	Pulmonary
QA	Quality assurance
QC	Quality control
r^2	Regression coefficient squared
SD	Standard deviation
SEM	Scanning electron microscopy
TB	Tracheobronchial
TCDD	Tetrachlorodibenzo-*p*-dioxin
TEF	Toxic equivalency factor
TEQ	Toxic equivalent
TOC	Total organic carbon
TWA	Time-weighted average
USGS	U.S. Geological Survey
WHO	World Health Organization
wt	Weight
µg	microgram
µL	microliter
µm	micrometer

PREFACE

Dioxins were discovered in ball clay in 1996 as a result of an investigation to determine the sources of elevated dioxin levels in two chicken samples from a national survey of poultry. The investigation indicated that the contamination source was ball clay added to chicken meal as an anti-caking agent. The purpose of this study is to evaluate another potential exposure scenario associated with ball clay, namely its use in ceramic art studios. This exploratory investigation makes preliminary exposure estimates that can be used to evaluate whether more detailed follow-up analyses are needed. Hypothetical dioxin exposure estimates were calculated using an assumption of dioxin levels in the ball clay based on measurements from other studies. The study was conducted during 2003 and 2004 by the National Center for Environmental Assessment with contract support provided by Battelle in Columbus, Ohio.

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1. INTRODUCTION AND BACKGROUND

Ball clay is a natural clay mined commercially in the United States, primarily in Kentucky, Tennessee, and Mississippi. A total of 1.21 million metric tons was mined in the United States in 2005. Its plasticity makes ball clay an important commercial resource for a variety of commercial uses. In 2005, it was used as follows: floor and wall tile - 40%, sanitary ware (sinks, toilets, etc.) - 25%, exports - 17%, ceramics - 11%, fillers, extenders and binders - 4%, pottery - 1.5%, and miscellaneous purposes - 1.9% (USGS, 2007).

Dioxins were discovered in ball clay in 1996 as a result of an investigation to determine the sources of elevated dioxin levels in two chicken samples from a national survey of poultry (Ferrario et al., 1997). The investigation indicated that soybean meal added to chicken feed was the source of the dioxin contamination. Further investigation showed that the dioxin contamination occurred when ball clay was mixed with the soybean meal as an anti-caking agent (Ferrario et al., 2000b; U.S. FDA, 2000). In 1997, the Food and Drug Administration (FDA) asked producers or users of clay products in animal feeds to cease using ball clay in all animal feeds and feed ingredients (U.S. FDA, 1997).

The purpose of this study is to characterize the possible dioxin exposures of artists using ball clay in ceramic art studios. This exploratory investigation makes preliminary exposure estimates that can be used to evaluate whether more detailed follow up analyses are needed. The limited resources available for this study required a strategy to base the analysis on existing data to the fullest extent possible.

Dioxin exposure is primarily a function of the dioxin concentration in the clay and an individual's level of exposure to the clay. Although studies in the literature provided information about dioxin levels in clay, no information could be found on clay exposure levels in ceramic art studios. Therefore, this study was designed to measure total clay exposures in a ceramic art studio. No dioxin measurements were made in this study, rather the dioxin levels in ball clay were assumed based on measurements from other studies. Three exposure pathways were evaluated: inhalation, dermal contact, and incidental ingestion. The evaluations involved measuring levels of clay particulates in air, clay residues on skin, and clay deposition on media representing food and beverages. These data provided a basis for estimating potential dioxin exposures and resulting doses, conducting an initial analysis of which exposure pathways contribute most to total dose, and evaluating how individual behaviors affect exposure/dose. Ultimately, the data helped develop distributions for input parameters for conducting a Monte

Carlo analysis to estimate how dioxin exposure/dose may vary across a wide population of artists.

An alternative way to evaluate dioxin exposures is by blood testing. While this provides a direct measure of dioxin exposure, it represents exposures from all sources, not just work in an art studio. Also, a blood study would not have provided any insights about how dioxin exposures may occur in an art studio. Normal background exposures vary widely and factors such as diet and age are known to have large impacts on dioxin body burden. Accordingly a blood study would require a large number of subjects with controls to reduce the effects of these factors. Also blood tests have very high analytical costs. On the basis of costs alone, blood testing was beyond the scope of this effort. The clay exposure testing done here provided a low cost way to explore the problem and gives future researchers an informed basis for deciding if blood testing or other types of follow-up work are needed.

Dioxin concentrations and exposures are presented in terms of toxic equivalents (TEQs). TEQs allow concentrations of dioxin mixtures to be expressed as a single value computed by multiplying each congener concentration by a toxicity weight (toxic equivalency factor or TEF) and summing across congeners. TEFs are expressed as a fraction equal to or less than 1 with 1 corresponding to the most toxic dioxin congener, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD). The TEQ data presented here are based on TEFs from the 1998 World Health Organization (WHO) recommendations (Van den Berg et al., 1998). In 2005, WHO updated the TEFs (Van den Berg et al., 2006). As discussed in Section 4, these updates had little impact on the literature values used here, so no adjustments were made.

The term “dioxins” is used in this study to refer collectively to the tetra- through octa-chlorinated dibenzo-*p*-dioxins (CDDs) and chlorinated dibenzofurans (CDFs) with chlorine substitutions in all of the 2,3,7,8 positions. This term is commonly defined to include the 12 co-planar pentachlorobiphenyls (PCBs) which also demonstrate dioxin-like toxicity. However, PCBs are not addressed in this study. PCBs have been shown to make up a small fraction of the total TEQs in a wide variety of background soils (U.S. EPA, 2007) and therefore are probably not important contributors to TEQs in ball clay.

2. APPROACH OVERVIEW

While working in a ceramics studio, artists may be exposed to dioxin-contaminated clay via three pathways: dermal contact, particle inhalation, and incidental ingestion. Exposure could also occur via open cuts or eyes and this possibility is discussed in Section 9 on uncertainty. The general strategy and procedures used to characterize each pathway are described below.

2.1. GENERAL STRATEGY

The site selected for this study was the Ceramics Area in Hopkins Hall at Ohio State University (OSU) in Columbus, OH. The Ceramics Area, housed in the basement of Hopkins Hall, has eight rooms, including classrooms, studios, a storage area, a glaze-mixing area, a clay recycling area, and a furnace room. This facility was selected because it offered a convenient location for assessing exposures during a variety of typical ceramic art activities.

The exposure measurements were carried out in two separate studies. The first study was conducted in April 2003 and the second in July 2004. The results of both studies have been combined in this report. Seven artisans and one nonartisan staff member in the OSU Ceramics Department were recruited to serve as subjects for the first study, and two additional artisans were recruited for the second study. An open solicitation was presented to the students and departmental staff, and the first volunteers were selected. The subjects included three males and seven females ranging in age from about 20 to 40 years. Approval for human subjects was obtained via the Battelle Institutional Review Board (IRB) and EPA. Upon approval by the Battelle IRB and EPA, OSU determined that review by their IRB was not necessary. The testing was conducted while the subjects conducted a variety of unscripted tasks, including clay mixing/preparation, sculpting, pottery wheel work, and molding.

To assess dioxin exposure levels, it is necessary to estimate dioxin levels in the various exposure media (i.e., clay used by the artists, dust particles suspended in the studio air, and dust settled onto surfaces). No actual dioxin measurements were made in this study. Rather, dioxin levels were estimated using literature-reported concentrations of dioxins in ball clay and information about the amount of ball clay in the clay mixtures used by the artists. Details about this procedure are discussed in Section 4.

A questionnaire was administered to subjects during the first study to gather information on their routines involving clay artwork. The questionnaire data are presented in Appendix A and summarized in Section 6.

2.2. CHARACTERIZATION PROCEDURES

The following procedures were used to characterize each exposure pathway.

2.2.1. Dermal Contact

Dermal contact with clay can occur via direct handling of the clay, deposition from the air onto exposed skin, transfer from surfaces, and splashing during wheel operations. The amount of clay on skin was measured using rinsing procedures. Additionally, surface wipes were collected in work areas to evaluate dermal exposures via transfers from surfaces. To further evaluate dermal exposure, a dermatologist examined the condition of the stratum corneum, the outermost layer of skin, before and after each subject worked with clay. The primary focus of this examination was to determine if any damage to skin may have occurred that would affect dermal absorption.

2.2.2. Inhalation

Both personal and area air-monitoring techniques were used to assess inhalation exposures. Personal air samplers provide data most representative of an individual's exposure because they sample the air in a person's breathing zone and reflect changes in concentration due to their movement. An area sampler provides a general indication of exposure for people in its vicinity and also can achieve lower detection levels. Both the personal and area-monitoring techniques provided particle size-selective data, so that the deposition site of the particles in the respiratory tract (nose/mouth, tracheobronchial airways, and alveolar region) could be determined.

Two types of personal air samplers were used: real-time and time-integrating. Similarly, two types of area air samplers were used: real-time and time-integrating. The real-time air samplers provided data on particle levels on a nearly continuous basis (every minute). The integrating samplers collected particles over the entire time period of a work activity, yielding a time-weighted average (TWA) concentration. In this sampling design, the real-time exposure monitoring was used to assess frequency, magnitude, and duration of peak exposures as well as TWA across the entire sampling time, while the integrating samplers provided information on average exposures.

2.2.3. Ingestion

Inadvertent ingestion of clay or dust can occur in several ways. Clay particles in the air can deposit on food or in beverages. Deposition onto surrogate food samples (a quartz filter was used to represent food and a beaker of water was used to represent a beverage, see Section 3.1.5

for further details) was measured to evaluate this pathway. Ingestion can also occur via transfers from hands to food or cigarettes and via transfers to the mouth resulting from wiping the hands or licking the lips. These possibilities were evaluated qualitatively through observations about individual behaviors. Finally, ingestion can also occur via particle deposition in the nose, mouth, and tracheobronchial airways; clearance to the throat; and swallowing. This process was evaluated using inhalation modeling (Appendix G).

3. SAMPLING METHODS

Methods used for collecting, preparing, and analyzing samples are described below.

3.1. SAMPLE COLLECTION

Samples were collected from personal air, area air, skin rinses, surface wipes, and surrogate food and beverages.

3.1.1. Personal Air Sampling

The Respicon model 8522 particle sampler (TSI Incorporated, Shoreview, MN) is a two-stage virtual impactor with a three-stage gravimetric filter sampler. The sampler sorts airborne particulate matter into three size ranges. Each size range is collected on a 37-mm glass fiber filter (GFF). The particle size collection ranges are as follows: stage 1, aerodynamic particle diameter (D_{ae}) < 4 µm; stage 2, 4 < D_{ae} < 10 µm; and stage 3, 10 < D_{ae} < 100 µm.

Before the start of sampling, three preweighed GFFs were removed from their protective polystyrene containers (47-mm Millipore petri slides) and loaded into the Respicon using nonmetallic filter forceps. A unique laboratory record book (LRB) identification number was assigned to each GFF during tare weighing, and this weight was recorded onto the sampling data sheet at that time. The Respicon was then assembled, and the total flow checker head was installed. A personal sampling pump (SKC model no. 224-PCXR4, Eighty Four, PA) was attached to the total flow head, and the flow rate through the Respicon was adjusted to 3.11 liters per minute (L/min) ± 2%, according to the manufacturer's specifications. All flows were verified by employing a calibrated National Institute of Standards and Technology (NIST)-traceable Buck calibrator (Model M5, A.P. Buck, Orlando, FL). After confirmation of the manufacturer's suggested flow rates at each stage of the sampler, the total flow checker was replaced with the standard (100 µm) inlet head. A nylon chest harness (TSI Incorporated, Shoreview, MN) was used to place the Respicon in each subject's breathing zone, approximately 15–20 cm below the chin. The personal sampling pump was attached to the subject's belt and connected to the Respicon. Sampling was initiated by starting flow through the Respicon and continued throughout a subject's entire work shift, typically 2 to 2.5 hours. The average sampling volume was 387 L. Following sampling, the pump was turned off, the Respicon was disassembled, and the filters were returned to their polystyrene petri dish containers for transportation back to the laboratory for gravimetric analysis. Quality control samples, such as field blank samples and matrix spike samples, were collected and analyzed for each sampling technique (see Section 3.2.3).

The personal DataRAM-1000 (pDR-1000, Thermo Electron Corporation, Franklin, MA) sampler was also used to measure personal particle exposure passively. No pump is required for this instrument; instead, the air surrounding the sampler circulates freely through the open sensing chamber by natural convection, diffusion, and background air motion. Particle concentrations are measured using a light-scattering (nephelometry) technique. This instrument responds optimally to particles with diameters in the range of 0.1 to 10 µm but will also respond to a lesser extent to larger diameter particles. Via internal calibration, the sampler converted particles/m^3 to mg/m^3 as final data units.

Before the start of sampling, the instrument sensor was zeroed by placing it in a resealable bag into which particle-free (filtered) air was pumped. All zero operations were performed successfully. To begin sampling, the instrument was clipped to the subject's waistline (on the belt or strap holding the SKC pump) and the unit was activated. The pDR-1000 collected data at 1 Hz and was programmed to record these data as 1-minute averages over the duration of the sampling period. At the conclusion of sampling (typically 2–2.5 hours), data logging was stopped and the instrument was turned off. The data were then uploaded to a personal computer using software provided by the manufacturer and an RS-232 serial port connection.

3.1.2. Area Air Sampling

To assess the particle size and concentration in the ceramic studio's air, a six-stage Delron cascade impactor (Delron Research Products, Powell, OH) was employed. Each stage filters out successively smaller particles so that the following particle sizes are collected in successive stages: >32 µm, 16–32 µm, 8–16 µm, 4–8 µm, 2–4 µm, and 0.5–2 µm; the final GFF collects all particles smaller than 0.5 µm in diameter. Particles accumulate on glass slides underneath each impactor orifice. To prevent particle loss due to bouncing, a small amount of vacuum grease was applied to each glass slide. The area coverage of the grease on the slide was determined by the approximate size of the impactor nozzle below which the slide was to be placed. Correct airflow rate through the impactor ensures that the correct particle sizes are collected on each stage. A carbon-vane pump (Gast Co., Benton Harbor, MI), with a critical orifice that provides a pressure drop of at least 430 mm of mercury, was used to ensure the flow rate of 24 L/min.

Before the start of sampling, preweighed glass slides were removed from their protective polystyrene petri slide containers and loaded into the impactor using clean forceps or tweezers. Unique LRB numbers, assigned to each slide during tare weighing, were recorded on sample data forms. The impactor tower was then assembled and flow was initiated to verify the required pressure drop. For each sample, the pressure drop was between 480 and 510 mm of mercury.

Flows were also verified using the Buck calibrator. Sampling times were approximately 2–2.5 hours, giving an average sample volume of approximately 2,900 L. Following sampling, the impactor was disassembled and all slides were returned to their respective petri dish containers for transportation back to the laboratory for gravimetric analysis.

The Climet CI-500 innovation laser particle counter (Redlands, CA) was a second sampling device used to measure area particle concentrations. In a manner similar to the pDR-1000, the Climet CI-500 measures particle number concentration using nephelometry. A self-contained pump sampled air at a constant flow rate of approximately 3 L/min. In the count mode, the Climet CI-500 measures particles in six particle size ranges: 0.3–0.5 µm, 0.5–1 µm, 1–2.5 µm, 2.5–5 µm, 5–10 µm, and >10 µm. The sampling frequency for the instrument is 1 Hz, and the data were logged as 1-minute averages. The particle counts were converted from particles/m^3 to mg/m^3 as final data units. The particle counts did not exceed the manufacturer's recommended maximum (200–250 counts/cm^3 at 3 L/min) at any time except for a few minutes during two of the sampling periods. No instrument zero or span checks were necessary. Following sampling, the data were uploaded to a computer using an RS-232 serial cable and software provided by the manufacturer. The Climet CI-500 was located in close proximity to the cascade impactor and generally very near the subject. For example, when the subject was working with clay at a wheel, the two air samplers were placed on the side of the wheel opposite the subject at a height and distance from the wheel similar to the subject's mouth and nose. The inlet to the Climet was oriented in a vertical direction.

3.1.3. Skin Sampling

The total skin area of hands, arms, face, feet, and legs was estimated using a combination of direct measurements and regression models based on body weight and height (U.S. EPA, 1997). The subject's exposed body parts were rinsed with a dilute soap solution (~2% soap in deionized [DI] water, by weight). Approximately 100–150 mL of the soap solution was used to rinse each exposed body part. After each body part was rinsed, the washbasin contents were transferred to a polypropylene bottle with small amounts of deionized (DI) water rinses. The bottle was labeled and sealed with a screw-top cap. The washbasin was then rinsed again, wiped out, and reused. Between the first and second studies, the procedures differed as described below.

April 2003. All subjects wore short-sleeved shirts, long pants, socks, and shoes. Therefore, the only exposed skin areas were the hands and forearms, and the rinsing was limited to these body parts. At three times during each subject's work session, the subject's exposed skin was examined for clay residue. When clay was observed visually, the affected areas of the subject's body were rinsed. Rinses were performed at approximately equally spaced intervals, and the last rinse usually coincided with the conclusion of the sampling period. The average of the three measurements was used to represent the session.

July 2004. Both subjects wore short-sleeved shirts, short pants, and sandals. Therefore, the exposed skin areas included the hands, arms, legs, and feet, and the rinsing was expanded from the first tests to include all of these body parts. The subjects' faces were also rinsed during these tests. Although no visible residues were apparent on the faces, this area was included for the sake of completeness.

The rinse samples were collected in a washbasin using a squirt bottle of soap solution while the subjects used their hands to gently wipe off the affected area. Rinses were conducted in the following manner:

- **Hands.** Moving downward from the wrist, the technician rinsed the residual clay off both sides of the artisans' hand; the residual clay from each hand was rinsed into separate containers and analyzed separately.
- **Arms.** Moving downward from the elbow, the artisans rinsed the residual clay from their arms.
- **Feet.** Moving downward from the ankle, the artisans rinsed the residual clay from their feet.
- **Legs.** Moving downward from the top of the exposed area of the legs, the artisans rinsed the residual clay from their legs.
- **Face.** The artisans rinsed the residual clay from their faces.

Skin rinse samples were collected at the close of each work session. In addition, if at any point during the work session the subject indicated the need to wash an exposed body part, it was rinsed into a sample container reserved for that body part.

3.1.4. Surface Wipe Sampling

A 20 cm by 20 cm horizontal surface near the subject's workspace was selected and cleaned with dilute soap solution before the subject began working with any clay. Wipe samples of this area were taken immediately after cleaning (to confirm that low levels were present before starting the work session) and at the end of the work session. The wipe sampling procedure consisted of the following steps. The selected area was wiped with 10 cm x 10 cm rayon gauze wipes wetted with ~5 mL isopropanol using the following procedure. The wipe was secured between the thumb and forefinger of one hand, and the surface was wiped five times in one direction using evenly applied pressure. The soiled side of the wipe was folded to the inside and, in an orthogonal direction, the surface was wiped five more times. This soiled side of the wipe was again folded to the inside and the wipe was placed into its prelabeled, resealable bag for transportation back to the laboratory for gravimetric analysis. The entire wiping process above was then repeated using one additional wipe.

3.1.5. Surrogate Food and Beverage

An 85-mm diameter quartz fiber filter and a 125-mL polypropylene jar filled with 100 mL DI water served as surrogates for food and beverage samples, respectively. Before clay work began, both were placed in a location where the artisan indicated he or she might normally place food or drink. In most cases, this location was away from the direct work area but still in the same room. However, occasionally clay workers placed food and beverage directly adjacent to their work. To begin sampling, the lid of the polycarbonate petri dish containing the food surrogate and the screw-cap lid on the beverage surrogate were removed. Following the conclusion of sampling, the lid to the petri dish was replaced and sealed with Teflon tape, and the polypropylene jar was secured for transportation back to the laboratory for gravimetric analysis.

3.2. SAMPLE PREPARATION AND ANALYSIS

Procedures used for sample preparation, analysis, and quality control are described below.

3.2.1. Filtration and Drying

To collect the clay rinsed from the subject’s skin during the skin rinse sampling procedure and the clay deposited into the surrogate beverage sample, the clay-liquid suspensions were filtered through a preweighed 85-mm diameter quartz fiber filter in a Buchner funnel using vacuum filtration. Any remaining clay in the sample container was rinsed with several small aliquots of DI water to ensure complete transfer of the clay to the filter. All filters from the vacuum filtration procedure were subsequently placed on clean 10-cm watch glasses and dried overnight at 100˚C. The gauze wipes for surface residues were dried in this fashion as well. No drying was required for the 37-mm Respicon filters or glass slides.

3.2.2. Gravimetric Analysis

The accuracy of the analytical balance (AT-20, Mettler-Toledo) used for all gravimetric analyses was confirmed daily with weights approved by NIST. The calibration weights ranged from 0.001 mg to 100 g. All 37-mm GFFs, 85-mm quartz fiber filter paper, 37-mm glass slides, and gauze wipes were conditioned in a temperature- and humidity-controlled balance room (temperature 22–23° C, relative humidity 46–56%) for a minimum of 24 hours before tare and final weights were recorded. For conditioning, the lid of the container holding the filter or slide was left slightly ajar, and the resealable bags containing the gauze wipes were left open. For both kinds of filters and glass slides, three separate weights were recorded to the nearest microgram. The weight was acceptable if the range of the three independent measurements was less than 10 µg. For gauze wipes, the three separate weights were recorded to the nearest tenth of a milligram and the acceptability criterion was that the range of the measurements be less than 1 milligram.

3.2.3. Quality Control Samples

At least one field blank sample was collected for each type of gravimetric sample, including the Respicon, cascade impactor, food and beverage, and surface wipe samples. Such samples were collected by transporting the sampling media to the field location and placing them into their respective sampling device or position for sampling. As soon as the medium was ready for sampling, it was collected as if the sampling time had come to a close and transported back to the laboratory for gravimetric analysis. The detection limits for the gravimetric measurements were determined by multiplying the standard deviation of the field blank net weights by 3. The detection limits for each type of gravimetric measurement were as follows: 0.0025–0.015 mg/m^3 for each stage of the cascade impactor, 0.878 mg/m^3 for each stage of the Respicon, 10.6 mg for the surface wipes, 0.6–1 mg for the food/beverage deposition samples, and 0.6–1.6 mg for the dermal rinse samples.

As a quality control check, the skin rinse, surface wipe, and food and beverage sampling and analysis methods were tested in a controlled laboratory setting. For the skin rinse method evaluation, approximately 3 g of clay (obtained from one of the artisan subjects) was handled carefully without dropping any until the entire sample was spread over the hands and forearms of a Battelle researcher. The skin rinse and analysis method described above was performed, and recoveries of 87 ± 3% of the clay applied were obtained. This compares favorably with Kissel et al. (1996), who obtained 93% recovery when rinsing wet soil from the skin of human subjects using a similar sampling method. Similarly, for the surface wipe method, approximately 1 g of clay was deposited onto a precleaned laboratory bench, the wipe method described above was performed, and recoveries of 94 ± 5% were obtained. For the food and beverage samples, approximately 50 mg of clay was added to those sampling matrices and recoveries of 90 and 95%, respectively, were obtained using the gravimetric analysis procedures described above.

4. DIOXIN CONTENT OF CLAY AND STUDIO RESIDUES

As discussed earlier, this study made no dioxin measurements in clays, dust residues, or other materials from the Ohio State University ceramics studio. Instead, the possible levels were estimated on the basis of other studies. A number of studies have measured dioxin levels in raw and processed ball clay. Raw clay is the clay as it comes out of the ground. Processed clays are the result of the initial processing, which is usually conducted at or near the mining site before shipping. This processing typically involves drying with hot air at 120˚C and pulverizing in a series of milling stages (Ferrario and Byrne, 2002). The following studies describe dioxin levels in raw and processed clay:

- **Ferrario and Byrne (2000, 2002).** Both papers present data for processed ball clay used at one ceramics manufacturer. The mean of seven samples of processed ball clay was 3,172 pg/g TEQ. Additional data are presented on dioxin levels in clay mixtures and fired products. The authors noted that dioxin levels in the dust samples collected at the facility were the same as those in the unfired clay mixtures.

- **Ferrario et al. (2000a).** This study compared the mean levels in eight raw clay samples from Mississippi (see Table 1) to the mean levels in four processed ball clay samples. This comparison showed that the processed clays had much lower levels of 2,3,7,8-TCDD and higher levels of 1,2,3,4,7,8-hexachlorodibenzo-*p*-dioxin (HxCDD), 1,2,3,4,6,7,8-heptachlorodibenzo-*p*-dioxin (HpCDD), and octachlorodibenzo-*p*-dioxin (OCDD) than the raw clay. The mean total TEQ of the processed clay (977 pg/g TEQ) was 37% lower than the raw clay (1,513 pg/g TEQ).

- **Ferrario et al. (2000b).** This study also presents the data for raw and processed clay described in Ferrario et al. (2000a). In addition, it presents dioxin levels in a variety of other types of clays and discusses the evidence of a natural origin for their presence.

- **Ferrario et al. (2004, 2007).** These studies collected processed ball clay directly from four art-supply retailers. All ball clay types sold by these retailers were purchased in 22.7 kg (50 pound) bags. One type of ball clay was sold by all four retailers, five types were sold by two of the retailers and seven types were sold by only one retailer. Thus a total of 21 bags representing 13 different types of ball clays were purchased and sampled. A ceramics expert confirmed that the most commonly used ball clays for making artware and pottery were represented in these samples. These data are summarized in Table 2.

Table 1. Raw ball clay dioxin concentrations

Congener	PCDD concentration (pg/g dry weight)			
	Range	**Median**	**Mean**	**Mean TEQ**
2,3,7,8-TCDD	253–1,259	617	711	711
1,2,3,7,8-PeCDD	254–924	492	508	508
1,2,3,4,7,8-HxCDD	62–193	134	131	13
1,2,3,6,7,8-HxCDD	254–752	421	456	46
1,2,3,7,8,9-HxCDD	1,252–3,683	1,880	2,093	209
1,2,3,4,6,7,8-HpCDD	1,493–3,346	2,073	2,383	24
OCDD	8,076–58,766	4,099	20,640	2
Total				1,513

TEQ = toxic equivalent

Source: Ferrario et al. (2000a).

Since the data from Ferrario et al. (2004, 2007) represented the types of clays most likely used in ceramic art studios, these data were selected as the most representative ones to be used in this study. Accordingly, it was assumed here that the dioxin TEQ levels in clay could range from 289 to 1,470 pg/g with an average of 808 pg/g. As shown in Table 2, the TEQs from this study were calculated on the basis of the WHO-98 Toxicity Equivalecy Factors or TEFs (Van den Berg et al., 1998). In 2005, WHO updated the TEFs (Van den Berg et al., 2006). These updates increased the TEF for OCDD from 0.0001 to 0.0003. None of the TEFs for the other six congeners used to estimate the ball clay TEQs were changed by the WHO update. The increase in the OCDD TEF would cause the overall average to increase by 6%. It was decided to use the TEQ estimates for ball clay as originally reported instead of updating it on the basis of the 2005 WHO TEFs. This was based on two reasons, first the change would have been relatively minor and second it would have complicated comparisons to exposure estimates which have not yet been updated on the basis of the new TEFs.

Table 2. Processed ball clay dioxin concentrations (pg/g)

	Average	Standard deviation	Median	Minimum	Maximum	WHO-TEF[a]	Avg TEQ
PCDDs							
2,3,7,8-TCDD	76	60	63.5	21.8	291	1	76.0
1,2,3,7,8-PeCDD	374	144	387	125	588	1	374
1,2,3,4,7,8-HxCDD	335	141	313	142	636	0.1	33.5
1,2,3,6,7,8-HxCDD	526	204	523	167	944	0.1	52.6
1,2,3,7,8,9-HxCDD	1,480	608	1,570	394	2,550	0.1	148
1,2,3,4,6,7,8-HpCDD	9,780	4,480	8,600	3,940	19,500	0.01	97.8
OCDD	254,000	88,200	233,000	118,000	471,000	0.0001	25.4
Total							
TCDD	1,450	606	1,600	412	2,370		
PeCDD	4,600	1,890	4,880	1,560	7,140		
HxCDD	13,500	5,710	12,800	4,800	21,900		
HpCDD	25,000	11,700	24,400	9,320	44,900		
Total TEQs[b]	808	318	771	289	1,470		808

[a]World Health Organization Toxic Equivalency Factors (WHO-TEFs) based on Van den Berg (1998)
[b]The overall average presented by Ferrario et al. (2007) is based on averaging the mean congener levels across samples. An alternative approach is to compute the average on the basis of the TEQ for each sample. This approach yields an average of 819 pg/g (SD = 303 pg/g). Similarly the median TEQ is 810 pg/g based on the individual samples. The minimum and maximum TEQ values are reported on the basis of the individual samples.
TEQ = toxic equivalent

Source: Ferrario et al. (2004, 2007).

All of these studies indicate that ball clay has relatively high levels of CDDs and very low levels of CDFs. Based on Ferrario et al. (2004, 2007), about 95% of the TEQs in processed clay are contributed by four congener groups: TCDDs (9%), pentachlorodibenzo-*p*-dioxin (PeCDDs) (46%), HxCDDs (28%), and HpCDDs (12%).

Artists commonly use a mixture of clays to achieve various physical properties and visual effects. The percentage of ball clay in the mixture can vary widely. The amount of ball clay in the mixtures used on days when the testing occurred ranged from 0 to 100% with an average of 21.5% (Table 3). Although 4 of the 10 subjects used mixtures containing no ball clay on the test days, on other days these subjects would likely use mixtures that do contain ball clay. This is because students are required to conduct a variety of projects, and some of these are better suited to using ball clay and others are not. Accordingly, it was assumed here that the ball clay portion of clay mixtures used by artists can range from 0 to 100% with an average of 20%. Furthermore, it was assumed that the dioxin levels in the non-ball clays were negligible. This is supported by Ferrario et al. (2000b), who analyzed 15 different mined clays and concluded their dioxin levels were significantly lower than levels in ball clay.

Table 3. Percentage ball clay in the clay mixtures used during this study

Subject	Percentage ball clay
1	0
2	27
3	48
4	0
5	20
6	0
7	0
8	15
9	100
10	5

Finally, it was assumed that the dusts suspended in the air and settled onto food or skin would have the same dioxin levels as the clay. Material other than clay may contribute to these dusts, further diluting dioxin concentrations. This possibility was evaluated using scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS). These techniques were applied to four types of samples:

- Blank GFF.

- Dust on a GFF collected from a storeroom at the Battelle Laboratory (not impacted by clay).

- Air particles on a Respicon GFF collected in the studio.

- Clay used by subjects.

SEM photographs and elemental spectra of samples associated with Subject 6 are shown in Figure 1. A visual comparison of the SEM photographs suggests that the particles on the Respicon filter appear to differ from those in the storeroom dust. Also, the spectra of the particles on the Respicon filters resemble clay more than those of storeroom dust. The clay samples and Respicon filter samples had high abundances of titanium, iron, and aluminum, which were not seen in the GFF blank or in the storeroom dust sample. Similar results were found for all eight subjects in the April 2003 tests, as shown in Appendix E. The analysis was not repeated in the July 2004 tests. These observations suggest that clay dominates the air particles collected in the studio. On this basis, it was assumed that the studio dust was dominated by clay and no further dilution factor was needed to adjust dioxin concentrations.

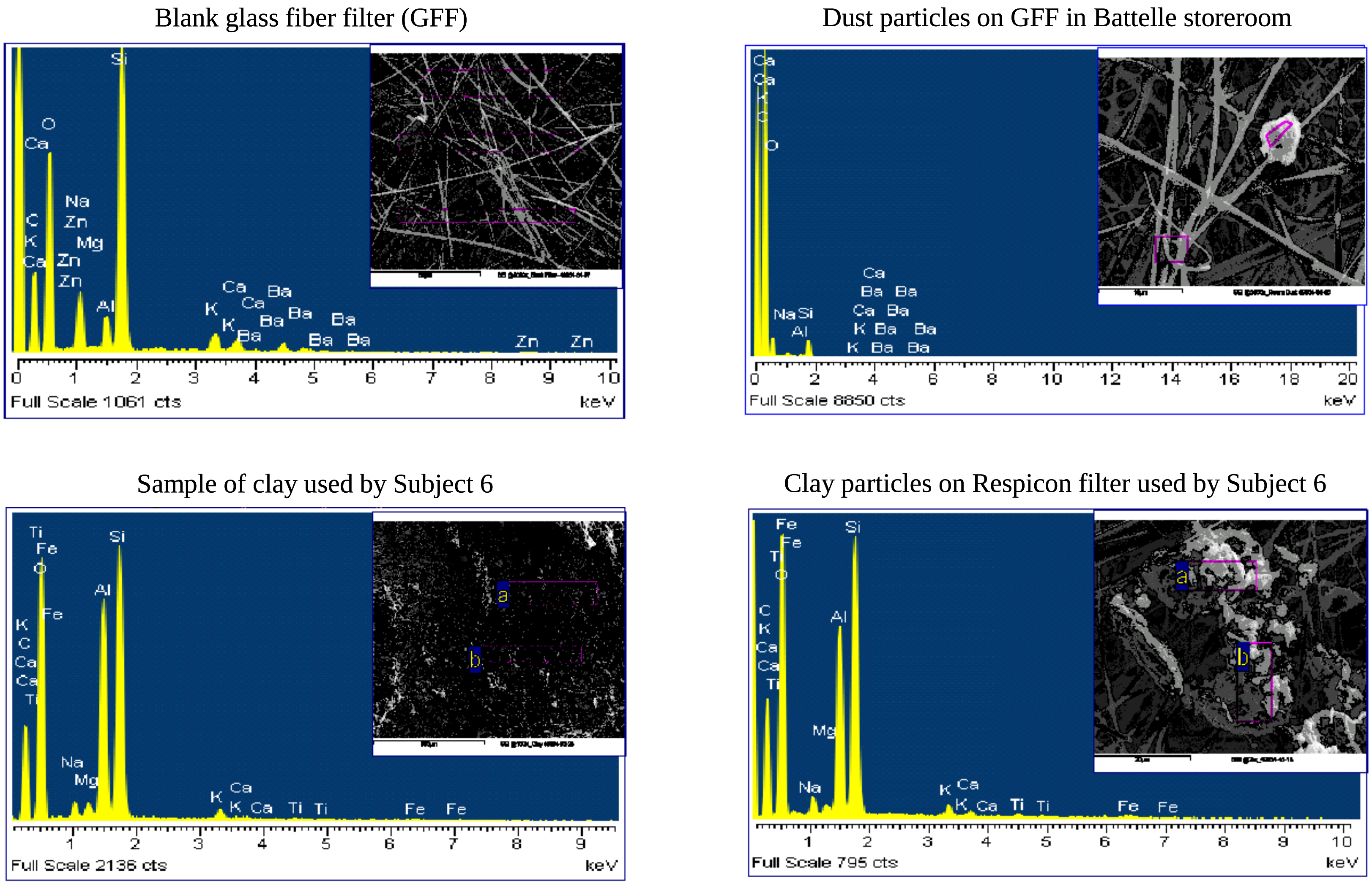

Figure 1. Scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) data.

5. DOSE ESTIMATION PROCEDURES

This section presents the procedures used to estimate the dioxin dose to artisans from all three routes of exposure: dermal contact, inhalation, and ingestion. Because the dermal dose is expressed on an absorbed basis, the dose by other pathways must also be expressed on an absorbed dose basis. This provides an equivalent basis for comparison and addition across pathways. All doses are presented as daily estimates. No adjustments are made for the frequency with which artists work with clay. Therefore, these dose estimates should be interpreted as the dose that could occur on a day that clay work is conducted, rather than as a long-term average.

5.1. DERMAL CONTACT

A fraction absorbed approach is used to estimate dermal absorption. This method has been widely used to assess dermal exposures to solid residues and is endorsed in current Agency guidance (U.S. EPA, 2004, 1992). Bunge and Parks (1998) have proposed an alternative approach based on a more mechanistic model. This model has had only limited testing and is not addressed in Agency guidance. Therefore, it was not chosen as the primary basis for this assessment, but Appendix I discusses how it could be applied to this situation. This new model suggests similar estimates of absorbed dose to those presented here using the traditional absorption fraction approach.

5.1.1. Estimating Particle Loading on Skin

As described earlier, rinsing procedures were used to determine the total amount of clay on exposed skin. This mass was divided by the exposed skin area to derive a loading in units of mg/cm^2.

5.1.2. Estimating Monolayer Load

The monolayer is the layer of particles immediately adjacent to the skin. According to the monolayer theory, the only significant dermal absorption comes from chemicals contained in this first layer (U.S. EPA, 2004, 1992). Experimental evidence supporting the monolayer theory has been published by Duff and Kissel (1996) and Touraille et al. (2005). To properly apply the dermal absorption fractions, it was necessary to determine whether residue loads on skin exceeded monolayer loads. The monolayer load for a specific soil can be estimated on the basis of the median particle size. Assuming spherical particles and face-centered packing, the monolayer loads can be calculated as follows (U.S. EPA, 2004):

$$L_{mono} = \rho \, d_p / 6 \qquad (1)$$

where:

L_{mono} = monolayer load (mg/cm^2)

ρ = particle density (mg/cm^3)

d_p = physical particle diameter (cm)

The average particle density of the processed clays analyzed by Ferrario et al. (2004) was 2.64 g/cm^3. Clays typically have very small particles relative to other components of soil. The U.S. Department of Agriculture (USDA) defines clays as having less than 2 µm diameter particles (Brady, 1984). The particle size specifications for a Tennessee ball clay is shown in Table 4 (Ceramics Materials Info, 2003). Reviewing the specifications for a variety of commercial ball clays, median particle sizes ranged from about 0.5 to 1.0 µm (Ceramics Materials Info, 2003).

Table 4. Particle size distribution of Tennessee ball clay

Particle diameter (µm)	20	10	5	2	1	0.5	0.2
% finer than	99	97	93	81	72	56	35

Source: Ceramics Materials Info (2003).

The particle sizes found in the studio air had median physical diameters ranging across subjects from 8 to 27 µm (this is derived from the mass median aerodynamic diameter [MMAD] range of 13 to 44 µm described in Appendix G and converted to physical diameters using the procedure in Appendix G, footnote 1). These airborne particles appear larger than what would be expected from the original clay product. This may be explained by the bonding of particles caused by the addition of water to the clay or the firing process, which fuses particles. Particles that accumulate on the skin primarily from air deposition are likely to resemble the air particles more than the original clay particles. Particles that transfer to skin primarily from direct handling of the clay should more closely resemble the original clay product than the airborne particles. Accordingly, the particle sizes of the clay residues on skin could vary widely, with medians ranging from 0.75 to 27 µm. For purposes of the central exposure estimates, the geometric mean of this range is assumed, i.e., 4.5 µm. This implies a monolayer load of 0.62 mg/cm^2. The uncertainty resulting from this assumption is discussed further in Section 9.

5.1.3. Estimating Fraction Absorbed

As discussed in U.S. EPA (1992), three teams of investigators have examined dermal absorption of TCDD from soil (Roy et al., 1990; Shu et al., 1988; Poiger and Schlatter, 1980). The Roy et al. (1990) data (also described in U.S. EPA, 1991) were selected as the best basis for estimating dermal absorption fractions applicable to the ceramics studio. This was because the test soil was most fully described allowing comparisons to the clay, and multiple exposure times were used allowing evaluation of how dose varies with time.

Roy et al. (1990) conducted a variety of experiments in which TCDD was applied to soil on human skin in vitro, rat skin in vitro, and rat skin in vivo. The experiments were conducted with both a low organic carbon soil and a high organic carbon soil. Ferrario et al. (2004, 2007) studied 21 samples of processed ball clay used in ceramics studios. They found that the organic carbon content of these samples ranged from 0.06% to 1.1% with a median and geometric mean of approximately 0.4%. This level is very similar to the level in the low organic carbon soil used by Roy et al. (0.45%). Accordingly, this discussion focuses on the Roy et al. results for the low organic carbon soil.

Roy et al. (1990) calculated the percentage absorbed at various times over the 96-hour experiment (Table 5). The second column shows the results for the human skin in vitro experiments. The percentage absorbed includes the amount measured in the skin at the end of the experiment. These values were adjusted in two ways. First, as recommended in U.S. EPA (1992), they were multiplied by the ratio of the percentage absorbed for rat skin in vivo (16.3%) to percentage absorbed for rat skin in vitro (7.7%). Second, they were adjusted to reflect the assumption that the absorption occurs exclusively from the monolayer. In the low organic carbon soil tests, Roy et al. (1990) used "Chapanoke" soil, which is composed of 15.1% sand, 68.2% silt, and 16.7% clay. Chapanoke soil has an organic matter content of 0.77% (0.45% organic carbon). Based on the USDA soil classification system, this composition is a silty loam. Silty loams have a median particle size of about 10 µm (Brady, 1984), which corresponds to a theoretical monolayer load of 1.3 mg/cm^2. Roy et al. applied a soil load of 6 mg/cm^2, exceeding the monolayer load by a factor of 4.6. Accordingly the percentage absorbed was also multiplied by this factor. The results of these two adjustments are shown in the third column of Table 5.

Table 5. Adjustments to Roy et al. (1990) dermal absorption data

Time (hr)	Percentage absorbed - human in vitro	Percentage absorbed - adjusted[a]	Percentage absorbed - best fit[b]
1	0.19	1.85	1.01
2	0.25	2.43	1.24
4	0.24	2.34	1.69
8	0.19	1.85	2.59
24	0.45	4.38	6.19
48	1.08	10.52	11.59
72	1.71	16.65	16.99
96	2.42	23.57	22.39

[a]These values were adjusted first by multiplying by the ratio of the percentage absorbed for rat skin in vivo (16.3%) to percentage absorbed for rat skin in vitro (7.7%) and second by multiplying by 4.6 to reflect the assumption that the absorption occurs exclusively from the monolayer.
[b]These values were derived using eq. 2 and converting to percent.

The Roy et al. (1990) data show a strong linear correlation between percent absorbed and time ($r^2 = 0.98$). The scatter plot for these data and the best fit line are shown in Figure 2. The equation for this line is as follows (converting percent to fraction):

$$AF_{dermal} = 0.00225t + 0.00787,\ t < 96hr \qquad (2)$$

where:

AF_{dermal} = dermal absorption fraction
t = time (hr)

This equation was adopted in this study for purposes of estimating dermal absorption of dioxin. The percentage absorbed values based on this equation are shown in the last column of Table 5.

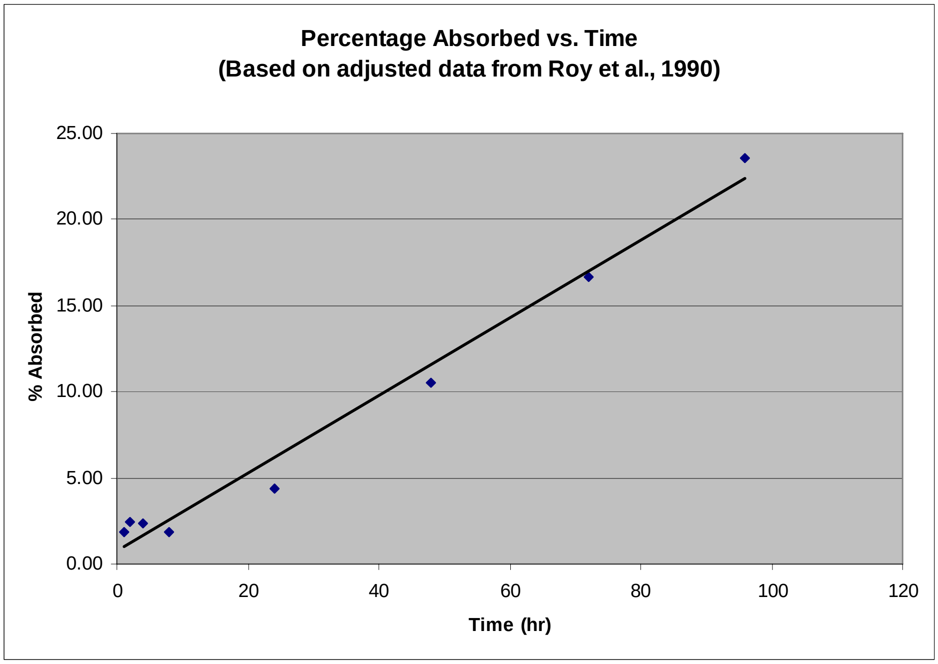

Figure 2. Scatter plot of adjusted absorption data versus time with linear trend line.

Source: Adapted from Roy et al. (1990).

5.1.4. Calculating Dermal Dose

The rinsing experiments indicated that clay loading exceeded the monolayer load in some, but not all, cases. The dermal absorption fractions presented above were applied to the measured loads where these were less than or equal to monolayer loads. At soil loadings greater than monolayer, the dermal absorption fraction was applied to only the monolayer load. Accordingly, the dose of dioxins absorbed through the skin of the artisan subjects during this study was estimated using the following equation for each body part and then summed:

$$D_{dermal} = SA\ L\ C\ AF_{dermal} \tag{3}$$

where:

D_{dermal} = dermally absorbed dose (pg TEQ/d)
SA = skin area exposed (cm^2)
L = daily clay loading on skin (measured or monolayer, whichever is less) (mg/cm^2-d)
C = dioxin concentration in clay (pg TEQ/g)
AF_{dermal} = dermal absorption fraction

5.2. INHALATION

The portion of particles that enter the respiratory tract through the nose or mouth (inhalability) depends mainly on particle size, route of breathing (through the nose or mouth), wind speed, and a person's orientation with respect to wind direction. Inhaled particles may be either exhaled or deposited in the extrathoracic (ET), tracheobronchial (TB), or pulmonary (PU) airway. The deposition of particles in the respiratory tract depends primarily on inhaled particle size, route of breathing, tidal volume, and breathing frequency (ACGIH, 2004; ICRP, 1994). Appendix G presents a detailed discussion of how to consider these factors and estimate the amount of particulate that deposits in various regions of the respiratory tract.

The absorbed inhalation dose is estimated as follows:

$$D_{inhalation} = D_r\ C\ AF_r\ (1g/1000\ mg) \tag{4}$$

where:

$D_{inhalation}$ = inhalation dose (pg TEQ/d)
D_r = dose of particles to region r of the respiratory tract (mg/d)
C = dioxin concentration on particles (pg/g)
AF_r = absorption fraction for region r of the respiratory tract

This equation is used to estimate the absorbed dose to the three regions of the respiratory tract (ET, TB, and PU) and then summed to derive total inhalation dose. In general, particles deposited in the ET and TB regions clear rapidly (within 1–2 days) to the throat and are swallowed. Accordingly, the absorption of dioxin from particles deposited in these regions is treated as if the particles had been ingested with an absorption fraction of 0.3 (U.S. EPA, 2003). The particles depositing in the PU region remain there a long time, and most of them are ultimately absorbed directly into the body (assumed absorption fraction of 0.8 based on U.S. EPA, 2003).

5.3. INGESTION

The ingestion dose is estimated by assuming that all particles deposited on the surrogate food and beverage samples are ingested. For both types of samples, the dose was calculated using the equation below:

$$D_{ingestion} = (F + B)\ C\ AF_{ingestion} \tag{5}$$

where:

$D_{ingestion}$ = ingestion dose (pg TEQ/d)
F = deposited clay on food (g/d)
B = deposited clay on beverage (g/d)
C = dioxin concentration in clay (pg TEQ/g)
$AF_{ingestion}$ = absorption fraction for ingestion

$AF_{ingestion}$ was assumed to equal 0.3 based on recommendations in U.S. EPA (2003) for ingestion of dioxin in soil. The ingestion of dioxin from inhaled particles is included in the inhalation dose as discussed above.

5.4. TOTAL DOSE

The total absorbed dose was estimated to be the sum of the dermal absorption, inhalation, and ingestion doses as shown below:

$$D_{total} = D_{dermal} + D_{inhalation} + D_{ingestion} \quad (6)$$

where:

D_{total} = total dose (pg TEQ/d)
D_{dermal} = dermally absorbed dose (pg TEQ/d)
$D_{inhalation}$ = inhalation dose (pg TEQ/d)
$D_{ingestion}$ = ingestion dose (pg TEQ/d)

6. QUESTIONNAIRE RESULTS

The complete questionnaire and all responses are presented in Appendix A. The questionnaire focused on characterizing each subject's work with clay in terms of frequency/duration, type of activity, clothing worn, and impact on skin. Table 6 summarizes the questionnaire results for the amount of time that the subjects spent working directly with clay. The subjects worked with clay, on average, for 30 hours per week and 38 weeks per year over a 6-year period. The times varied widely, however, reflecting the types of students involved. A student obtaining an advanced degree in ceramics is likely to work with clay daily over many years. In contrast, a student who takes a pottery class to fulfill a general education requirement is likely to experience similar exposures, but only for 1–3 hours per day over the duration of the class (9 months or less).

Table 6. Questionnaire questions on duration and frequency of subject's clay work

Question (n = 8)	Mean (SD)	Median	Max	Min
Approximately how many hours per week do you work with clay?	30 (21)	23	70	10
Approximately how many weeks per year do you work with clay?	38 (10)	38	52	20
How long (years) have you been doing clay work with this level of intensity?	6 (8)	3	24	1

Table 7 summarizes the participants' answers to several questions about their clay work. Some of the questions address the types of clothing worn, how often the subjects wash their hands, and whether the subjects could correlate any skin health effects with working with clay. All eight subjects answered that they have dry skin because of the clay work. In general, the subjects wash their hands soon after working with clay, their face and arms within a few hours, and the rest of their body within 24 hours. The responses indicated that one subject gets a rash when using the wheel for throwing, another subject has nasal congestion due to clay work, and another subject's fingernails do not grow well.

Table 7. Questionnaire questions about clay work

Question (n = 8)	Summary of answers (number of subjects with similar answers)
What type of clay artwork do you do?	Hand building/sculptural work (7), throwing on wheel (3), mixing clay and maintenance work (1)
What types of clothing do you wear while you work?	In general, long sleeves and pants in cool weather and short sleeves and pants or shorts in warm weather; both closed-toe shoes and sandals are worn at times
What areas of skin typically are exposed to the clay while you work?	Always face and hands; arms, legs, and feet when exposed
In relation to the time you complete working with clay, when do you wash parts of your body that have been exposed to clay?	Soon after: hands (8), arms (1), face (1) Within a few hours: arms (2), face (6) Within 24 hours: face (1), rest of body (4)
How do you wash your skin after you work with clay?	Soap and water or just water (8)
Do you correlate any skin health issues with how much you work with clay? If yes, what?	Dryness (8), rash on hands when using wheel (1), nasal congestion (1), fingernails do not grow well (1)

7. COMPARING EXPOSURES ACROSS SUBJECTS

In this section, a hypothetical dioxin dose is estimated for each subject and used to evaluate which pathways and activities contribute most to total dose. This is done by assuming that each subject uses clay with the same level of dioxin. More specifically, it is assumed that each subject uses a clay mixture with 20% ball clay and that the ball clay contains 808 pg TEQ/g (these are typical values as discussed in Section 4). Accordingly, the dioxin levels in the clay were assumed to be 20% of 808 pg TEQ/g or 162 pg TEQ/g. This concentration was also assumed to apply to inhaled dust and dust settled onto food. A variety of other factors were also held constant across subjects to facilitate this analysis:

- **Exposure duration.** The questionnaire results presented in Section 6 indicate a median weekly time for clay work of 23 hours. Assuming a 5-day work week, this would correspond to about 4 hours/day. This value was applied to all subjects.

- **Monolayer load.** The monolayer load varies depending on particle size but is assumed here to be 0.62 mg/cm^2 for all subjects. This is based on the geometric mean of the range of possible median particle sizes, i.e., 0.75 to 27 µm (see Section 5.1 for further discussion of this issue).

- **Dermal absorption fraction.** This will depend on exposure time, as discussed in Section 5.1. The time that the skin is exposed to clay will vary with individual behaviors and body parts. Some body parts (such as hands and faces) are likely to be washed more frequently than others (such as feet, legs, and arms), resulting in longer exposure times. The questionnaire data collected during this study (see Section 6) suggest that the artists generally wash their hands soon after working with clay, wash their faces and arms within a few hours, and wash the rest of their body within 24 hours. Accordingly, the exposure time for feet and legs was assumed to be 24 hours, and the absorption fraction corresponding to 24 hours was applied (6.2%). The exposure time for hands, arms, and face was assumed to be 4 hours with a corresponding 1.7% absorption.

- **Ingestion absorption fraction.** This was set to 0.3 based on recommendations in U.S. EPA (2003) for ingestion of dioxin in soil.

- **Inhalation absorption fraction.** This was set to 0.3 for ET and TB regions based on the assumption that the area is rapidly cleared to the gastrointestinal tract. It was set to 0.8 for the PU region based on recommendations in U.S. EPA (2003) for inhalation of dioxin in air.

The hypothetical dioxin dose for each subject is calculated using the constant values described above and their individual exposure conditions (e.g., dust level in air, clay load on skin, clay load on food). The dose estimates are considered to be hypothetical because they are based on assumed dioxin levels in the various exposure media rather than on studio-specific measurements. Section 8 presents an analysis of the possible variability in dose resulting from a range of dioxin levels in clay, ball clay mixtures, and exposure factors (Monte Carlo simulations).

This section first addresses each pathway separately (dermal contact, inhalation, and ingestion) and then addresses total dose. Individual exposures vary widely, and it is important to consider the subject's activity and clothing in evaluating the results. Table 8 is provided as a reference for this purpose with summaries of each participant's activities and clothing.

7.1. DERMAL CONTACT

As described in Section 5.1, the mass of clay rinsed from the skin was used to estimate clay loadings on the skin for each exposed body part. The rinsing data are presented in Appendix H. Section 5.1 also explains that the skin loading is compared to the monolayer load, and the absorption fraction is applied to the lower amount. The dermal absorption estimate for each subject is shown in Table 9. Subjects 1 through 8 wore clothing that limited their exposures to only hands and arms (although arm exposure was detected on only Subjects 1 and 6). The estimates for Subjects 9 and 10 include hands, arms, legs, and feet because they wore clothing allowing exposure to these areas. All subjects could have had exposure to the face, but this was evaluated only for Subjects 9 and 10. Pictures of the clay residues on skin are shown in Appendix B. Table 9 shows that 5 of the 10 subjects had skin exposures exceeding the monolayer. The absorbed dose ranged from 0.41 to 20.80 pg TEQ/d with a mean of 3.37 pg TEQ/d (SD = 6.18).

The relationships between the activities of the subjects and their dermal exposure, as presented in Table 9, are discussed below:

- **Wheel work (Subjects 6 and 9).** This activity led to the highest dermal exposures. The high exposures were caused by the close proximity of the subjects to the wheel, the splashing of wet clay onto their bodies, and the use of both hands to mold the clay. The total dermal dose for Subject 9 was about 6 times greater than that for Subject 3, resulting primarily from their clothing difference. Both had similar hand and arm exposure, but Subject 9 had high exposure to legs and feet and Subject 6 had no exposure in these areas.

Table 8. Artisan activities of each subject

Artisan/staff (minutes sampled)	Description of activity	Clothing
Test 1, April 2003		
Subject 1/male (153 min)	Wedged clay on a wedging board to remove air from the clay before kneading and shaping clay by hand. Used a wooden press to press the clay into flat, approximately 2.5 cm thick sheets. Also, pounded semi-dry clay into balls, placed in ball mill for smoothing rough edges.	Short-sleeved shirt, long pants, socks, shoes
Subject 2/male, nonartisan staff (84 min)	Poured powdered components into large mixer for clay manufacture while wearing dust mask and while the dust removal system was operational. Weighed out portions of clay, and bagged and stored them. Subject moved to gas kiln room, where he cut blocks, built the kiln up a bit, and vacuumed. Finally, subject used compressed air to clean the dust off himself.	Short-sleeved shirt, long pants, socks, shoes
Subject 3/female (124 min)	Subject wedged clay and covered a prefabricated mold with clay using her hands to mold and shape the clay.	Short-sleeved shirt, long pants, socks, shoes
Subject 4/female (121 min)	Subject cut pre-wedged and formed blocks of clay into 5 cm thick pieces, loaded the blocks into a pneumatic press, pressed a pattern into each and cut blocks to the proper shape, and then stacked the finished pieces to be fired.	Long-sleeved shirt (rolled up), long pants, socks, shoes
Subject 5/male (136 min)	Subject hand rolled clay into 60 cm long "snake-like" cylinders, which he then hand-formed into conical pots.	Short-sleeved shirt, long pants, socks, shoes
Subject 6/female (123 min)	Subject threw a variety of clay items, including a pitcher, a vase, pots, and bowls on the pottery wheel.	Short-sleeved shirt, long pants, socks, shoes
Subject 7/female (124 min)	Subject wedged, rolled, cut, and hand-built a variety of items.	Short-sleeved shirt, long pants, socks, shoes
Subject 8/female (138 min)	Subject wedged, rolled, shaped, cut, and hand-built large pieces of clay and placed them on a mold.	Short-sleeved shirt, long pants, socks, shoes
Test 2, July 2004		
Subject 9/female, five sessions (295–476 min)	Subject threw a variety of clay items, including plates, bowls, vases, and cups, on the pottery wheel.	Short-sleeved shirt, short pants, sandals
Subject 10/female, three sessions (406–438 min)	Subject sculpted detailed designs into clay tiles and plaques; also chipped small bits of excess clay off pieces of art that had already been fired.	Short-sleeved shirt, 3/4 length pants, sandals

Table 9. Hypothetical estimates of dermal dose

Body part	Clay load on skin (mg/cm²)[c]	Skin area (cm²)[e]	Fraction uncovered	Absorbed dioxin dose (pg TEQ/day)[a,b,d]
Subject 1				
Hands	0.38	970	1.0	1.00
Arms	0.15	2,406	0.5	0.49
Total				1.50
Subject 2				
Hands	[2.01]	970	1.0	1.65
Subject 3				
Hands	0.51	865	1.0	1.2
Subject 4				
Hands	0.17	855	1.0	0.41
Subject 5				
Hands	[2.61]	1,005	1.0	1.71
Subject 6				
Hands	[9.25]	790	1.0	1.34
Arms	[2.99]	2,005	0.6	2.04
Total				3.38
Subject 7				
Hands	0.26	785	1.0	0.57
Subject 8				
Hands	[1.90]	715	1.0	1.21

Table 9. Hypothetical estimates of dermal dose (continued)

Body part	Clay load on skin (mg/cm^2) [c]	Skin area (cm^2)[e]	Fraction uncovered	Absorbed dioxin dose (pg TEQ/day)[a,b,d]
Subject 9				
Hands	[10.12]	857	1.0	1.45
Arms	[1.50]	2,265	0.75	2.88
Lower legs	[0.72]	2,161	1.0	13.44
Feet	0.26	1,151	1.0	2.99
Face	0.03	374	1.0	0.03
Total				20.80
Subject 10				
Hands	0.20	783	1.0	0.42
Arms	0.04	2,271	0.9	0.22
Lower legs	0.11	2,095	0.1	0.23
Feet	0.03	1,109	1.0	0.30
Face	0.04	368	1.0	0.04
Total				1.22

[a]Absorption = skin load (mg/cm^2-day) × skin area (cm^2) × fraction uncovered × dioxin concentration in clay (pg TEQ/g) × 10^{-3} mg/g × absorption fraction.
[b]All calculations assume dioxin concentration in clay = 162 pg TEQ/g and absorption fraction is 6.19% for feet and legs, and 1.69% for hands, arms, and face.
[c]All bracketed loads exceed monolayer of 0.62 mg/cm^2 and were reduced to this value in absorption calculation.
[d]Results from Subjects 1 through 8 are based on one work session, from Subject 9 are based on average of five sessions, and from Subject 10 are based on average of three sessions.
[e]Skin area is for total body parts; for two-sided parts, it is the sum of right and left sides.
TEQ = toxic equivalent

- **Mixing (Subject 2).** Subject 2 was involved in the mixing and handling of dry clays and furnace/kiln maintenance during the work session. This activity produced relatively large hand loadings.

- **Wedging and molding (Subjects 1, 3, 4, 5, 7, and 8).** Wedging clay involves kneading and hitting clay against a tabletop to purge air pockets from the clay. During the wedging process, the clay is firm and dry as compared with clay used on the wheel. This activity produced a wide range of hand loadings (from 0.17 to 2.61 mg/cm^2).

- **Sculpting (Subject 10).** This involved sculpting activities on dry clay. At times, fine detailing tools were used that involved very little contact with the clay, resulting in low hand loading.

Table 10 shows the percent contribution to the dermal dose by body part for Subjects 9 and 10. Subjects 9 and 10 were tested in July 2004 and wore summer clothing, which allowed exposure to their legs and feet. Leg and foot exposure accounted for 79% of the total dose for Subject 9 and 44% of the total dose for Subject 10. This reflects the relatively large surface areas and higher absorption fraction (due to longer exposure time) for these parts. The uncovered portion of Subject 10's lower legs was only 10%, so the leg contribution to total dose was much less than that of Subject 9. Facial exposures were low, accounting for only 0.1–3% of total dose.

Table 10. Percent contribution to dermal dose by body part

Body part	Percentage of dose	
	Subject 9 (wheel)	Subject 10 (sculpture)
Hands	7	34
Arms	14	18
Legs	65	19
Feet	14	25
Face	0.1	3

7.1.1. Clay Loads on Surfaces

The horizontal surfaces in ceramic art studios can have high dust loads resulting from air deposition. Most clay on the hands of artisans probably results from direct contact with clay, but some could also result from contact with surfaces. In the interest of exploring this issue, wipe samples were collected from the work surface of each subject. These results are shown in Table 11. The surface dust loads ranged from 0.2 to 7 mg/cm^2, which are high compared with dust loads on floors in residences (i.e., 0.005 to 0.7 mg/cm^2) (Lioy et al., 2002). The efficiency of transfers from surfaces to hands will vary depending on the type of surface, type of residue, hand condition, force of contact, etc. Rodes et al. (2001) conducted hand press experiments on particle transfer to dry skin and measured transfers with central values of about 50% from hard surfaces. Several of the ratios of hand loads to surface loads given in Table 11 exceed 50% by a wide margin. Subject 6 was working on a wheel and clearly had hand loads resulting from direct contact with clay. Similarly, Subjects 5 and 8 had very high hand loads that must have resulted from direct clay contact. The other subjects had ratios ranging from 0.05 to 0.30, which are in the range that could result from surface transfers. Observation of the subjects indicated that almost all contact with the work surface also involved some contact with the clay. Therefore, the hand residues are most likely derived from a combination of direct clay contact and transfers from surfaces.

Table 11. Comparing clay loads on surfaces to clay loads on hands

Subject	Clay loading on surface (mg/cm^2)	Clay load on hand (mg/cm^2)	Ratio of hand load to surface load
1	7.002	0.38	0.05
2	NA	2.01	NA
3	2.966	0.51	0.17
4	0.572	0.17	0.30
5	0.774	2.61	3.4
6	0.238	9.25	38.9
7	1.206	0.26	0.22
8	0.419	1.90	4.5

NA = Nonartisan subject was not working at a surface during sampling, so this type of sample was not collected.

7.1.2. Dermatologist Report

The dermatologist did not diagnose any serious skin health problems among the subjects. Small abrasions and common skin conditions such as dryness and cracking, as the subjects reported on the questionnaires, were noted, but changes in these conditions could not be detected based on before and after observations.

7.2. INHALATION

Estimating the inhalation dose involved measuring particle concentrations in air and modeling deposition to various regions of the respiratory system. Classroom exposures were not estimated.

7.2.1. Particle Levels in Air

As described in Section 3, four different sampling techniques were used during the April 2003 tests to measure clay particle concentrations in air: two personal monitors and two area monitors. The data from all four devices are shown in Appendixes C and D. The Respicon personal air sampler normally would have been the best indicator of individual exposures, but the blanks were high, resulting in a high detection limit and a high frequency of nondetects in the data. Instead, the cascade impactor was chosen as the best indicator of daily exposure. Although this is an area sampler, it was located near the subjects and the subjects were generally stationary during the test. Thus, it should have been a reasonable indicator of individual exposures. Also, the cascade impactor uses deposition collectors and gravimetric techniques to estimate air concentrations; consequently, it is a more direct measurement technique than the other two instruments (pDR-1000 and Climet), which use light scattering to estimate particle concentration. These optical devices provide a nearly continuous readout of concentration levels, making them better suited to evaluating short-term fluctuations in particle levels rather than long-term concentrations.

Only the cascade and Climet monitors were used in the July 2004 tests. The instruments were located even closer to the individuals, i.e., within 30 cm of their breathing zones. The data were used in a fashion consistent with the April 2003 tests, i.e., daily exposures were based on the cascade data and the Climet was used to evaluate short-term fluctuations.

Table 12 presents the air data for each subject on the basis of the cascade measurements. The MMADs were estimated by fitting the data to log-normal distributions (see the discussion in Appendix G). Table 12 indicates that the range for total particulate matter is 0.084 to 0.99 mg/m^3. Note that the upper end of this range is less than the Occupational Safety and Health Administration (OSHA) standard for total particulates of 15 mg/m^3 (OSHA, 2004). Subject 3's concentration was the highest because students were cleaning the floor near the area samplers (see the discussion below). Subject 9’s concentration was the lowest as a result of a relatively low activity level during the testing. Subject 5’s concentration was also low, likely because a steady breeze entered through an open window in the room in which sampling was occurring. All of the other subjects had fairly similar concentrations.

Table 12. Particle concentrations in air and mass median aerodynamic diameter (MMAD) based on cascade impactor

Subject	MMAD (µm)	Total concentration (mg/m^3)
1	26.9	0.35
2	44.6	0.47
3	18.5	0.99
4	25.0[a]	0.37
5	25.0[a]	0.13
6	20.2	0.61
7	13.0	0.51
8	26.7	0.64
9	32.6	0.084
10	16.0	0.24

[a]Nondetects prevented calculation of the MMAD for these subjects; they were assumed equal to the average over the remaining first eight subjects.

The two subjects using wheels (Subjects 6 and 9) had very different air exposures. Because a great deal of water is used to moisten clay during wheel molding (the clay was saturated with water and a pan of water was placed directly next to the artisans for their use), this setting would not be expected to produce much clay dust, which was observed for Subject 9. Subject 6, however, had fairly high air levels. Subject 6 was located near a classroom that, as discussed below, had high activity levels. Therefore, this subject’s high air levels may have been associated more with the classroom activities than the wheel activities.

Figure 3 shows the plot of concentration versus time (based on the Climet CI-500 area particle counter) for Subject 3, who worked in an area designated for graduate student work adjacent to a large classroom. Approximately 50 minutes into the sampling session, about 20 students from the adjacent classroom began sweeping and wiping down the surfaces. This activity continued for approximately 15 minutes and generated a significant cloud of dust. As shown in Figure 3, particle levels began rising at about 50 minutes, peaked sharply at 60–70 minutes, and declined to low levels at about 80 minutes.

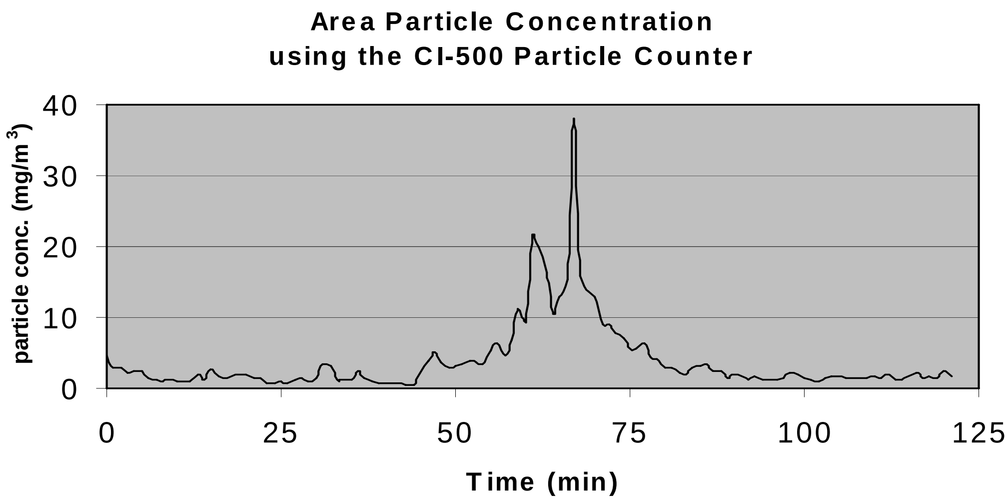

Figure 3. Real-time particle concentration for Subject 3 using the CI-500 particle counter.

During two of Subject 10's sculpture work sessions, a small dog was present. The dog's movement disturbed dust on the floor of the ceramics studio and, in turn, increased the particle concentration. Figures 4 and 5 are the real-time traces for the Climet monitor for the sculpting work sessions during which the dog was present. The dog was present for the entire first sculpting work session. This was reflected in the relatively constant variation in the particle concentration throughout the work session. During the second sculpting work session, the dog did not arrive until 138 minutes into sampling. Note the increase in overall particle concentration and increase in variability of particle concentration after arrival of the dog. The presence of a dog in the studios and classrooms is not likely to be a common occurrence, especially during the regular school year. Therefore, the particle concentrations during the work sessions when the dog was present (1 and 2) were not used to estimate the exposures for this subject. It should be noted, however, that pets, which may be present in many ceramic art studios, can have a large influence on the suspended dust levels and spread dust to other areas.

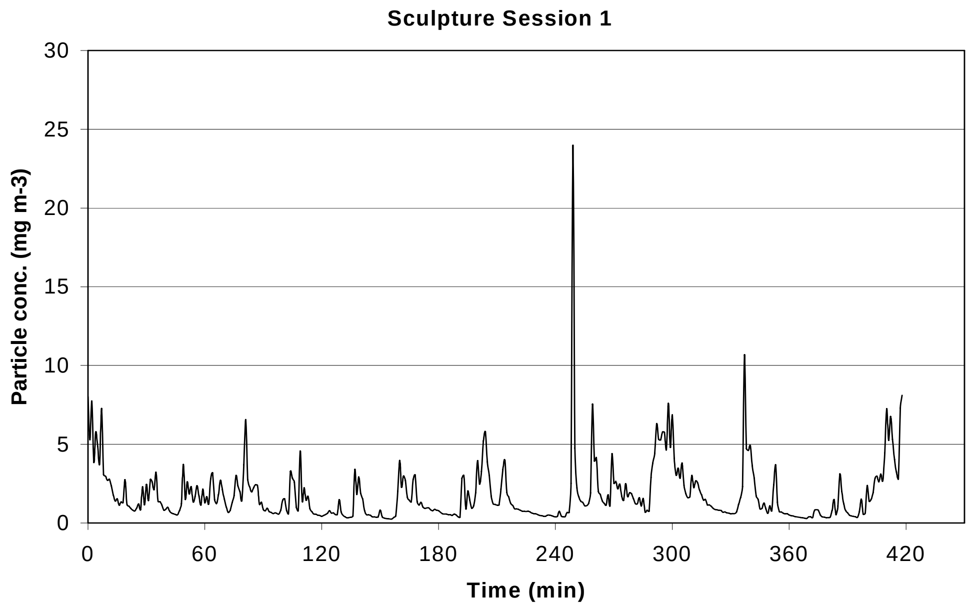

Figure 4. Sculpture session 1 with dog present.

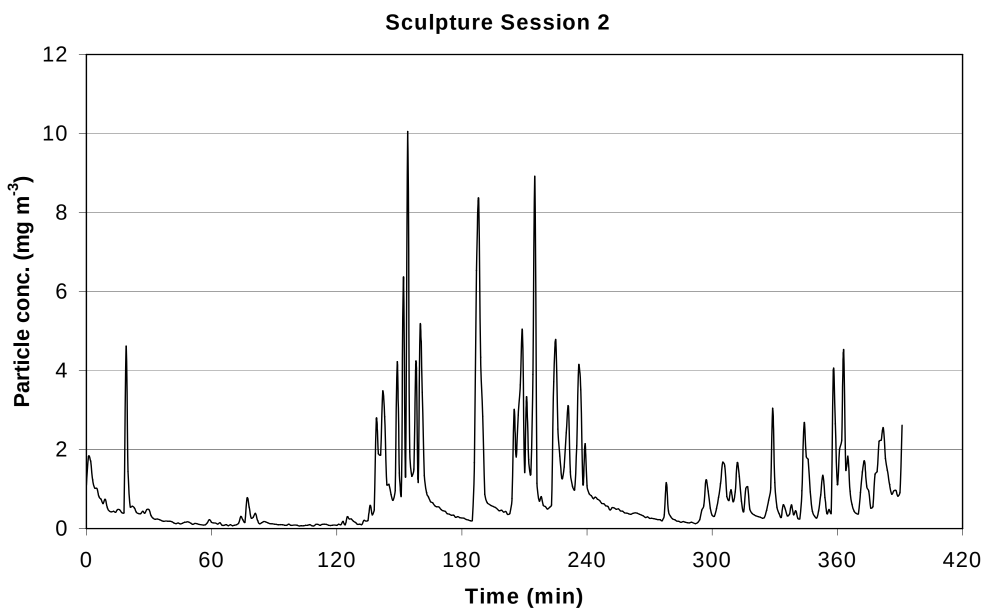

Figure 5. Sculpture session 2 with dog present.

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7.2.2. Inhalation Dose

Table 13 shows the absorbed dose in various regions of the respiratory system for all 10 subjects. The total inhalation doses ranged from 0.006 to 0.09 pg TEQ/d with an average of 0.04 pg TEQ/d. Most particle deposition was found to occur in the extrathoracic region. The modeling to support these estimates is presented in Appendix G.

Table 13. Hypothetical estimates of inhalation dose

Subject	Absorbed dose (pg TEQ)[a]			
	ET[b]	TB[b]	PU[c]	Total
1	0.032	0.001	0.003	0.035
2	0.033	0.001	0.003	0.036
3	0.082	0.002	0.010	0.094
4	0.028	0.001	0.002	0.031
5	0.012	0.000	0.001	0.014
6	0.054	0.001	0.004	0.059
7	0.049	0.001	0.006	0.057
8	0.048	0.001	0.003	0.052
9	0.005	0.000	0.001	0.006
10	0.022	0.001	0.002	0.025

[a]Dose calculated using procedures in Appendix G for nasal breathing; subject exposure concentrations from Appendix D; 4-hour exposure duration and dioxin concentration of 162 pg TEQ per gram clay.
[b]Absorption fraction of 0.3 assumed, since these regions rapidly clear into the gastrointestinal tract.
[c]Absorption fraction of 0.8 assumed, in part, due to slow particle clearance from this region.

TEQ = toxic equivalent; ET = extrathoracic; TB = tracheobronchial; PU = pulmonary

The inhalation exposure estimates assume that no respiratory protection was used. Generally this was true, however, Subject 2 used a dust mask while pouring powdered clay into a mixer for clay preparation. This reduced his inhalation exposures relative to levels reported here.

7.2.3. Classroom Exposure

Estimating student exposures in a classroom setting was not an objective of this study. However, some insight on this issue can be gained from the data for Subjects 1, 3, and 6. These subjects performed their clay activities adjacent to the undergraduate classroom during times when undergraduate classes of 20–25 students were participating in clay-related activities. The area particle samples collected for these subjects are generally representative of the inhalation exposure of students in those classes. As discussed above, students in this class swept the floor during Subject 3's testing period, producing elevated particle concentrations for about 30 minutes.

7.3. INGESTION

The ingestion dose was calculated by assuming that all deposited material on the surrogate food and beverage samples was ingested. As Table 14 shows, clay deposition onto the food and beverage samples reached detectable levels in only 5 out of 16 total samples. The deposition amounts for the nondetects were assumed to equal half the detection limit. The resulting ingestion doses ranged from 0.03 to 0.1 pg TEQ/d. The field technicians did not observe hand-to-mouth activities for any of the subjects. Also, none of the subjects ate food or smoked without first washing the clay from their hands. No deposition samples were collected for Subjects 9 and 10.

7.4. TOTAL DOSE

Table 15 lists the hypothetical estimates of total dioxin dose derived by summing across exposure pathways for each subject. The total doses ranged from 0.49 to 20.81 pg TEQ/d with an average of 3.45 pg TEQ/d. Table 16 shows the percentage contribution of each exposure pathway to the total dose of each subject. Dermal absorption is the major contributor to total dose for all subjects, exceeding 78% for all subjects. Ingestion and inhalation contribute similar amounts, generally in the range of 1–10%.

Table 17 shows the dose estimates by activity. The highest total doses were associated with wheel activities.

Table 14. Clay deposition and hypothetical estimates of ingestion dose

Subject	Clay deposited onto food (mg)	Clay deposited into beverage (mg)	Ingestion dose (pg TEQ/day)[a,b]
1	0.71	0.66	0.07
2	<DL	<DL	0.03
3	<DL	<DL	0.03
4	<DL	0.72	0.05
5	<DL	<DL	0.03
6	<DL	<DL	0.03
7	1.66	<DL	0.1
8	1.50	<DL	0.09

[a]Ingestion dose (pg TEQ) = (deposited clay on food (mg) + deposited clay on beverage (mg)) × dioxin concentration in clay (pg TEQ/g) × absorption fraction × (1 g/1,000 mg).
[b]All calculations assume dioxin concentration in clay = 162 pg TEQ/g, absorption fraction = 0.3, all deposited clay is ingested, and nondetects were set equal to half the detection limit.

TEQ = toxic equivalent; DL = Detection limit (0.60 mg).

Table 15. Hypothetical estimates of total dioxin dose (pg TEQ/day)

Subject	Estimated dioxin dose (pg TEQ/day)			
	Inhalation	Ingestion	Dermal absorption	Total
1	0.035	0.07	1.50	1.61
2	0.036	0.03	1.65	1.72
3	0.094	0.03	1.20	1.32
4	0.031	0.05	0.41	0.49
5	0.014	0.03	1.71	1.75
6	0.059	0.03	3.38	3.47
7	0.057	0.1	0.57	0.73
8	0.052	0.09	1.21	1.35
9	0.006	NM	20.80	20.81
10	0.025	NM	1.22	1.25
Mean (SD)	0.041 (0.025)	0.05 (0.03)	3.37 (6.18)	3.45 (6.15)
Median	0.036	0.04	1.36	1.48
Minimum	0.006	0.03	0.41	0.49
Maximum	0.094	0.10	20.80	20.81

TEQ = toxic equivalent; NM = not measured; SD = standard deviation

Table 16. Percent contribution to total dioxin dose

Subject	Percentage of dose		
	Inhalation	Ingestion	Dermal absorption
1	2.2	4.4	93.4
2	2.1	1.7	96.2
3	7.1	2.3	90.7
4	6.3	10.2	83.5
5	0.8	1.7	97.5
6	1.7	0.9	97.4
7	7.8	13.8	78.4
8	3.9	6.7	89.5
9	0.0	NM	100.0
10	2.0	NM	98.0

NM = not measured

Table 17. Dose estimates by activity

Activity	Subject	Inhalation dose (pg TEQ/day)	Ingestion dose (pg TEQ/day)	Dermal dose (pg TEQ/day)	Total dose (pg TEQ/day)
Wedging and molding	1	0.035	0.07	1.50	1.61
	3	0.094	0.03	1.20	1.32
	4	0.031	0.05	0.41	0.49
	5	0.014	0.03	1.71	1.75
	7	0.057	0.1	0.57	0.73
	8	0.052	0.09	1.21	1.35
Mixing	2	0.036	0.03	1.65	1.72
Wheel	6	0.059	0.03	3.38	3.47
	9	0.006	NM	20.80	20.81
Sculpting	10	0.025	NM	1.22	1.25

NM = not measured; TEQ = toxic equivalent

8. MONTE CARLO SIMULATION OF THE EXPOSURE DATA

Section 7 presented hypothetical dose estimates for each subject, assuming that all were using typical amounts of ball clay with average dioxin levels. In this section, Monte Carlo simulations are used to explore the doses that could occur in a broad population of artists with a wide range of behaviors using ball clay with differing levels of dioxin.

The general strategy for selecting input value distributions was as follows. The distribution of skin surface areas across adults in the general population was assumed to be log-normal with mean and standard deviation from the *Exposure Factors Handbook* (U.S. EPA, 1997). Similarly, the dioxin concentration in clay was assumed to have a log-normal distribution with mean and standard deviation from Ferrario et al (2004, 2007). The rationale for choosing log-normal distributions was that physiological parameters and environmental media concentrations are commonly found to have these types of distributions. The distributions were truncated at the minimum and maximum data points to eliminate the chance that some simulation trials could use unreasonable values. The remaining exposure factor parameters were based on observations from this study. These were generally assumed to have triangular distributions with ranges based on minimum and maximum values and peaks based on means. The rationale for choosing a triangular distribution was that (1) the small sample sizes associated with the study observations prevented fitting the data to standard distributions and (2) it reflected the likelihood that a central value would occur most often. In some cases (e.g., clay load on face), only two data points were available and a uniform distribution was assumed. The distributions assumed for all input variables are listed in Table 18.

Crystal Ball 7 software was used to conduct 1,000 trial simulations. For each simulation trial, a set of parameter values was obtained by randomly sampling the parameter distributions as listed in Table 18 and then computing the dioxin dose. The dose was calculated using the equations presented in Section 5. All simulation trials first select a set of values for the dioxin concentration in ball clay, the fraction of ball clay in the blend used by the artist, and the exposure duration. These are shown as general parameters in Table 18. The simulation then calculates the dose from the dermal, inhalation, and ingestion pathways, as discussed below:

- **Dermal.** The simulation was designed to first select a total body surface area from a log-normal distribution. Subsequently, skin surface areas for individual body parts were calculated by multiplying the total surface area by the average percentage of total surface area. These percentages were obtained from U.S. EPA (1997): hands, 5.2%; arms, 14%; legs, 31.8%; feet, 6.8%; and face, 2.5% (assumes face area equals one-third of head area). This approach ensures that simulation trials have realistically matched body part areas. Since the body part

Table 18. Monte Carlo simulation input parameters and sampling distributions

Parameter	Distribution	Basis
General parameters		
Dioxin concentration in ball clay (pg TEQ/g)	Log-normal (mean = 808, SD = 318)	Ferrario et al. (2004, 2007) (n = 21); truncated at range limits
Fraction of ball clay in blend	Triangular (0, 0.2, 1.0)	Data in this study (n = 10)
Exposure duration (hr/d)	Triangular (1, 4, 10)	Judgment and data from this study (n = 8)
Dermal absorption parameters		
Total body surface area (cm^2)	Log-normal (mean = 18,000, SD = 37.4)	*Exposure Factors Handbook* (U.S. EPA, 1997); truncated at range limits (n = 32)
Clothing selector	Uniform (0, 1.0)	Judgment and data from this study (n = 8)
Clay load on hand (mg/cm^2)	Triangular (0.1, 3.0, 10)	Range and mean based on observations from this study (n = 10)
Clay load on arm (mg/cm^2)	Triangular (0.04, 0.35, 3.0)	Data in this study (n = 4)
Clay load on leg (mg/cm^2)	Uniform (0.1, 0.70)	Data in this study (n = 2)
Clay load on feet (mg/cm^2)	Uniform (0.03, 0.3)	Data in this study (n = 2)
Clay load on face (mg/cm^2)	Uniform (0.03, 0.04)	Data in this study (n = 2)
Ingestion parameters		
Clay load on food (mg)	Triangular (0.3, 0.7, 1.66)	Range and mean based on observations from this study (n = 8)
Clay load on beverage (mg)	Triangular (0.3, 0.5, 0.72)	Range and mean based on observations from this study (n = 8)
Inhalation parameters		
Particle concentration in air (mg/m^3)	Triangular (0.08, 0.44, 0.99)	Range and mean based on observations from this study (n = 10)
Median particle size (µm)	Triangular (13, 25, 45)	Judgment and data from this study (n = 10)
Lung parameters	Male, 30%; female, 70%	Male/female split based on data in this study (n = 10)
Fraction of time engaged in light vs. moderate exertion.	Uniform (0, 1.0)	Judgment
Breathing type	Oronasal, 13%; nasal, 87%	Brown (2005)

area calculations give total areas, a fraction unclothed was used to reduce this to the exposed area. These fractions were based on four clothing scenarios as shown in Table 19. These clothing scenarios were based on questionnaire responses and judgment about typical apparel for a moderate climate. A clothing scenario was selected randomly for each simulation trial according to the time fractions shown in Table 19. Distributions were also assumed for the clay loads on skin. These were assumed to be spread uniformly over the entire unclothed area. As discussed in Section 5.1, dermal absorption was assumed to be limited to the monolayer that was held constant at the median value of 0.62 mg/cm^2 (the impact of changing this value is discussed as an uncertainty issue in Section 9). Finally, the absorption fractions (as presented in Section 5.1) were applied to derive the absorbed dose from exposed body parts and then summed to derive total dermal dose.

Table 19. Clothing scenarios based on questionnaire responses

Clothing scenario	Time fraction	Fraction unclothed		
		Arms	Legs	Feet
Long-sleeved shirt, long pants, shoes	0.2	0	0	0
Short-sleeved shirt, long pants, shoes	0.6	0.67	0	0
Short-sleeved shirt, short pants, shoes	0.1	0.67	0.67	0
Short-sleeved shirt, short pants, sandals	0.1	0.67	0.67	1.0

- **Inhalation.** The inhalation dose was calculated using the procedures summarized in Section 5.2 and presented in detail in Appendix G. Distributions were used to represent the variability in total particulate concentration in air and median particle size (see Table 18). Breathing was assumed to be either oronasal (13%) or nasal (87%), based on Brown (2005). Inhalation parameters (see Appendix G) were based on an average female for 70% of the trials and an average male for 30% of the trials. The rate of breathing was determined by the fraction of time engaged in light versus moderate exertion. These fractions were varied randomly from 0 to 1.0 using a uniform distribution. Depositions to various parts of the respiratory system were modeled as described in Appendix G, multiplied by the absorption fraction, and summed to derive the total inhalation dose.

- **Ingestion.** The variability in ingested dose was simulated using distributions for the levels of clay in the food and beverages as shown in Table 18. As discussed in Section 5.3, all deposited material was assumed to be ingested.

Two Monte Carlo simulations were conducted. The first simulation was designed to evaluate the influence of clay use only. Accordingly, it was conducted using the distributions for dioxin concentration in the clay and the fraction of ball clay in the blend used by the artists. All other inputs were held constant at their central values. The summer clothing scenario was used (i.e., short-sleeved shirt, short pants, sandals). This simulation produced a mean total dose of 39 pg/d, median of 33 pg/d, and 90th percentile of 73 pg/d. These results are best compared to the hypothetical dose estimate for Subjects 9 and 10 (presented in Section 7) because they wore summer clothing matching the simulation assumption. Subject 9 had a dose estimate of 21 pg/d, corresponding to about the 30th percentile of the simulation. Subject 10 had a dose of 1.5 pg/d, corresponding to about the 2nd percentile of the simulation. This simulation suggests that clay choice alone can account for a wide range of exposures with the potential to elevate exposures above the hypothetical estimates for the 10 subjects.

The second simulation used the distributions for all parameters as shown in Table 18. This simulation produced a mean total dose of 16 pg/d, median of 8 pg/d, and 90th percentile of 37 pg/d. The standard deviation exceeds the mean indicating that the results have a wide spread as shown in Figure 6. The hypothetical dose estimates of most subjects would have corresponded to low percentiles of this simulation except Subject 9 (80th percentile). Table 20 shows the simulation results for each pathway. The simulation means for each pathway exceeded by 3 to 4 times the means of the hypothetical dose estimates for the 10 subjects. As observed during the field study, the ingestion and inhalation doses are much smaller than the dermal dose. The frequency diagram for total dose is shown in Figure 6. This figure shows a highly skewed distribution with a peak around 3 pg TEQ/d and a long tail to the right extending to about 70 pg TEQ/d. A detailed report showing all inputs and outputs for this simulation is presented in Appendix F.

A sensitivity analysis was performed using the Crystal Ball 7 software. Each input parameter was evaluated using contribution to variance and rank order correlation (Figures 7 and 8). These analyses showed that clothing selected contributed most to variance (37.9%), followed closely by fraction of ball clay in blend (37.7%), dioxin concentration (16.6%), and exposure duration (5%).

Overall, the simulation suggests that higher exposures than those reflected in the hypothetical dose estimates of the 10 subjects may occur. This results from the skewed input distributions, which generally have long right-hand tails. Also 6 of the 10 subjects had hand exposure only, and the simulation uses a range of clothing that will result in more skin exposure in most trials.

Table 20. Descriptive statistics of dioxin doses from ball clay use, based on a Monte Carlo simulation

Pathway	Mean	Standard deviation	Median	90th Percentile
Dermal dose (pg TEQ/d)	15.5	22.91	7.92	36.15
Ingestion dose (pg TEQ/d)	0.14	0.10	0.11	0.28
Inhalation dose (pg TEQ/d)	0.12	0.13	0.08	0.27
Total dose (pg TEQ/d)	15.76	23.01	8.12	36.63

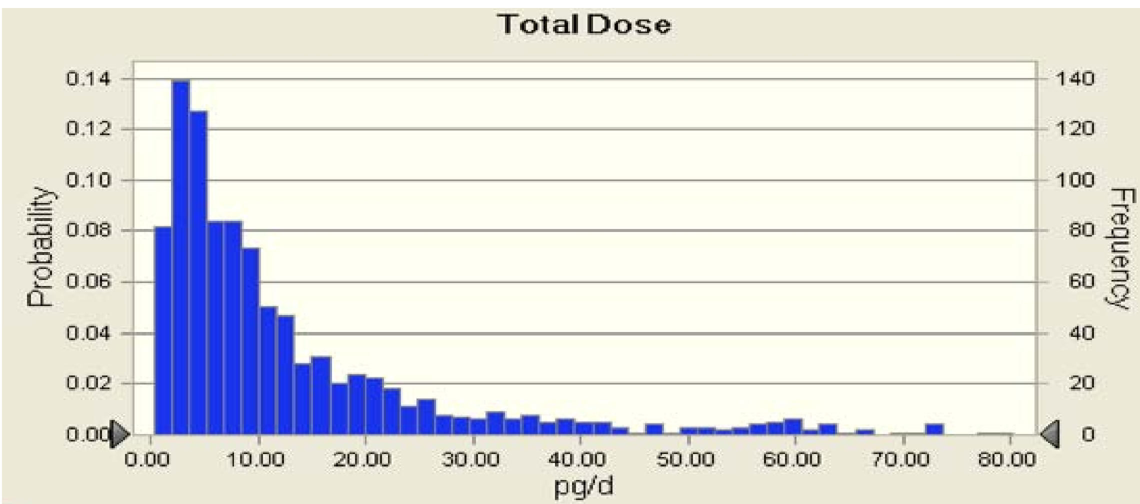

Figure 6. Frequency distribution of total dose (pg TEQ/day) based on Monte Carlo simulation.

Many of the input distributions used in this simulation were based on very limited data or judgment. A number of the distributions were based on data from this study, and the degree to which the study subjects represented a broader population of artists is unknown. Similarly, the degree to which the studio conditions observed in this study represent a broader set of studios is unknown. The simulation should be interpreted as a preliminary indication of how to extrapolate the study results to a broader population of artists.

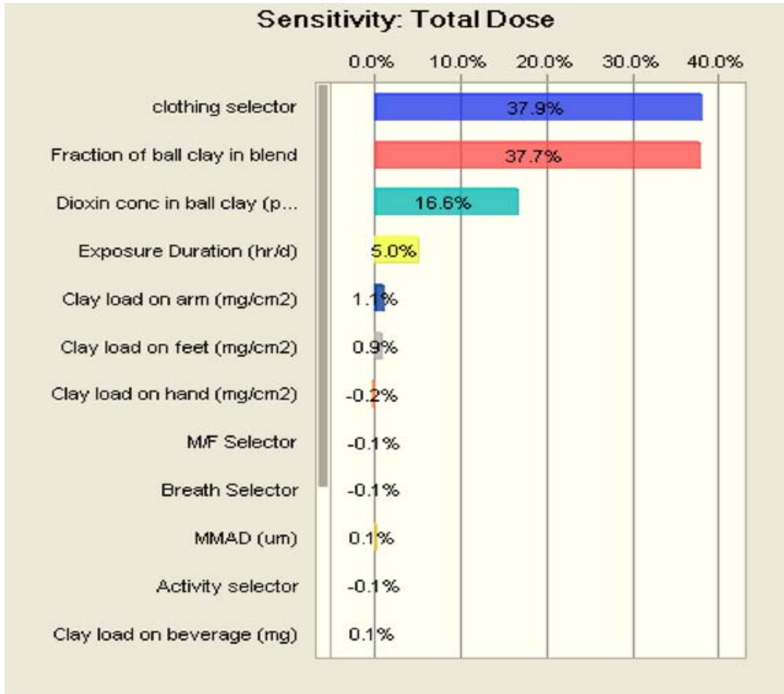

Figure 7. Sensitivity analysis based on percent contribution to variance.

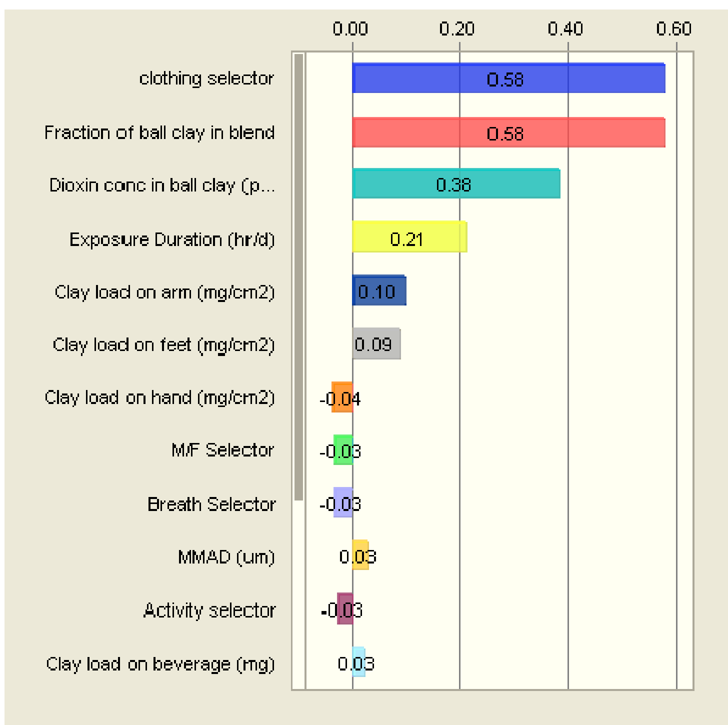

Figure 8. Sensitivity analysis based on rank correlation.

9. UNCERTAINTY

This section discusses general uncertainty issues and uncertainties related to the three exposure pathways: dermal, inhalation, and ingestion.

9.1 GENERAL UNCERTAINTY ISSUES

The sensitivity analyses showed that the dioxin concentrations in clay and the fraction of ball clay used account for a large part of the overall variance in the exposure estimates. Thus it is important to consider the uncertainty in the assumptions regarding these two parameters.

The dioxin levels in ball clay were assumed on the basis of the study by Ferrario et al. (2004, 2007). An important uncertainty issue is whether the ball clay sampled by Ferrario is representative of the ball clay used in the studio and by the broader community of ceramic artists. Ferrario et al. (2004, 2007) explained that the major mining companies market a total of 32 ball clay products of which 13 were sampled. Although marketing data were not available to do true statistical sampling, a ceramics expert confirmed that the most commonly used ball clays were included in this study. The samples were collected from 22.7 kg (50 pound) bags in the same form as delivered to ceramic studios. Four of the 21 samples analyzed by Ferrario et al. matched exactly the primary type of ball clay used in the OSU ceramics studio.

As explained earlier, ceramic artists use a wide range of clay blends with ball clay contents ranging from 0 to 100%. The hypothetical dose estimates were based on the assumption of 20% ball clay in the blend, which is the average fraction used by the 10 subjects in this study. It is unknown how representative this is of the wider population of ceramic artists. The ball clay fraction assumption may also affect other exposure factors. For example, it could affect how much clay adheres to skin. Soil adherence to skin has been shown to be influenced by moisture content and particle size. Ball clay is similar to other clays in terms of these properties. The primary way that ball clay is unique from other clays is its high plasticity. It is not known how this property would affect skin adherence.

9.2. DERMAL EXPOSURE UNCERTAINTIES

A fraction absorbed approach is used to estimate dermal absorption based on current Agency guidance. As discussed in Section 5.1, this method has acknowledged weaknesses, but the uncertainties are difficult to assess. Appendix I presents an alternative approach using a more mechanistic model. This model predicts an absorbed dose that is similar to the fraction absorbed approach. The mechanistic model has had limited testing, and it is not yet clear whether it provides more reliable estimates.

The exposures in the studio are caused by clay, but the dermal absorption fraction is derived from soil experiments. An important uncertainty issue is whether clay has properties that differ significantly from soil and consequently make the soil-derived absorption estimates invalid for clay. The soil used by Roy et al. (1990) was 16.7% clay. This fraction of the soil should have properties similar to those of the studio clay. The organic carbon content of the clay is approximately the same as that of the low organic soil used by Roy et al. In terms of particle size, clays typically have lower particle sizes than soil and would be expected to more strongly sorb organic contaminants (e.g., dioxins) as compared with normal soils, all other factors being equal. As discussed in Section 5, commercial ball clay specifications report a median particle size of about 0.75 µm, which is smaller than that of the Roy et al. soil (median diameter of about 10 µm). The particle sizes measured in the studio air had median diameters ranging from 8 to 27 µm, which are larger than those of the soils used by Roy et al. This may be explained by the bonding of particles caused by the addition of water to the clay or the firing process, which fuses particles. Thus, it appears that the particle size of the soil used by Roy et al. falls within the range present in the studio.

The studies on dermal absorption of dioxin from soil by Roy et al. and other investigators have exclusively used TCDD. It is important to consider whether results for TCDD can be extrapolated to the other dioxin congeners found in clay. As mentioned previously, the compounds of concern in the clay are the tetra- through octa-CDD congener groups, as listed in Table 21. This table indicates that molecular weight and the octanol-water partition coefficient (K_{ow}) increase with chlorine substitution. Molecular weight and K_{ow} have been identified as key chemical properties affecting dermal absorption (U.S. EPA, 1992). These properties also relate to how tightly bound chemicals are to soils and their release kinetics. The higher chlorinated congeners would be released from soils more slowly and permeate skin more slowly than TCDD. Thus, use of TCDD experiments to represent the penta - octa dioxin congeners found in clay probably leads to some overestimates of dermal absorption, but it is uncertain to what degree.

A related question is whether TCDD-derived dermal absorption values can be applied to TEQs. As shown in Table 21, only about 9% of the TEQ in processed clay is derived from TCDD. The TEFs used to determine TEQs discount the hepta- and octa- congeners much more than the tetra- and penta- groups. The overestimates of dermal absorption for the higher chlorinated congeners due to their higher molecular weights and K_{ow} values will be compensated to some extent by the large discounts during the TEQ calculation and thus make extrapolation of dermal absorption data from TCDD to TEQs more reasonable.

Table 21. Physical properties of dioxin congeners and concentration in processed clay

Congener	Molecular weight	Log K_{ow}[a]	Concentration in processed clay[b] (pg/g)	Concentration in processed clay[b] (pg TEQ/g)	% of total TEQ
TCDD	322	6.1 to 7.1	76	76	9
PeCDD	356.4	6.2 to 7.4	374	374	46
HxCDD	390.9	6.85 to 7.8	2,341	234	28
HpCDD	425.3	8.0	9,780	97.8	12
OCDD	459.8	8.2	254,000	25.4	3
Total				808	

[a]U.S. EPA (2000)
[b]Average values from Ferrario et al. (2004, 2007)

The amount of chemical that is dermally absorbed has been shown to be related to skin thickness and whether the skin is dead or alive (U.S. EPA, 1992). Skin thickness varies across body parts and across individuals. No information was found that could be used to account for these factors in this analysis.

As discussed in Section 5.1, the monolayer calculation is also an important source of uncertainty for the dermal absorption estimates. The monolayer load is estimated on the basis of the median particle size and assumption of ideal packing. Actual monolayers will be composed of a mix of sizes with complex packing that could result in loadings higher or lower than this theoretical estimate. It is also uncertain how to best characterize the size distribution of particles on the skin. The particles in the original clay product have a median particle size of about 0.75 µm, and the airborne particles have medians ranging from 8 to 27 µm. The particles on the skin could more closely resemble either the airborne particles or the clay particles, depending on the deposition mechanism. Accordingly, particle sizes of the clay residues on skin could vary widely, with medians ranging from 0.75 to 27 µm. For purposes of the central exposure estimates, the geometric mean of this range was assumed, i.e., 4.5 µm. This implies a monolayer load of 0.62 mg/cm^2. The monolayer loads corresponding to the upper and lower ends of the particle size range are 0.1 to 3.7 mg/cm^2. This uncertainty is dampened in the dose estimate as a result of the assumption that absorption occurs from only the monolayer. This dampening is especially strong for low-exposure subjects. For example, the dose estimates for Subject 4 (who had the lowest dermal exposure) corresponding to the low and high ends of the monolayer load range would be 0.23 and 0.41 pg TEQ/day. Thus, a 37-fold variation in monolayer load resulted in only a 1.8-fold variation in dose. The dampening is less (but still significant) for Subject 9 (who had the highest dermal exposures). For this subject, the doses corresponding to the low and high ends of the monolayer load range would be 4.1 and 34.2 pg TEQ/day, respectively.

Another source of uncertainty in the dermal absorption estimates concerns the condition of the skin. Some of the artists reported dryness and cracking of skin due to clay activities. These conditions were observed by the dermatologist, but correlation with clay activities could not be confirmed. Wheel operations involve work with wet clay which would hydrate the skin. The abrasive nature of this work could also reduce the thickness of the stratum corneum which is considered the primary barrier to permeation (U.S. EPA, 1992). It is possible that these conditions would allow more dermal permeation than normal intact skin. However, any increased permeation would be limited to the surface areas associated with the damaged skin. Exposure could also occur through the eyes where absorption would likely be greater than intact skin. This would be limited to particles that contact the eye surface which is probably minimal.

9.3. INHALATION UNCERTAINTIES

Data from the cascade sampler were used to estimate inhalation exposures. These data were considered to be the most reliable because no samples were below detection limits and the sampler uses a direct measurement method. The cascade, an area sampler, was located as near the subject as possible but normally would not represent an individual's exposure as accurately as a personal air monitor. Unfortunately, the data from the Respicon personal monitor were dominated by nondetects and could not be used. The limited Respicon data that were above detection limits generally indicated higher levels than the cascade, suggesting that personal exposures may have been higher than those detected by the area monitor. Accordingly, use of the cascade data may have resulted in underestimates of inhalation exposures.

9.4. INGESTION UNCERTAINTIES

The only ingestion pathway quantitatively evaluated in this study was direct ingestion of clay deposited from the air onto food items. The measured deposition onto surrogate food/beverage samplers may not match that of actual foods/beverages. Also, other pathways of ingestion may occur. For example, clay could be transferred from hands directly to food.

Although this transfer was not observed in this study, it could be a fairly common occurrence and has the potential for significant transfers to handheld food items (e.g., sandwiches, chips, cookies). Clay ingestion could also occur from wiping the mouth or licking the lips. The maximum ingestion levels estimated in this study involved about 2 mg of clay. This appears to be low when compared to the 50 mg/day adult soil ingestion rate specified as a default assumption in EPA guidance (U.S. EPA, 1997, 1989). This value is for residential scenarios and includes both outdoor soils and indoor dusts. While it is logical that dust ingestion alone would be less than ingestion of both soil and dust, a residence is likely to be much less dusty than a ceramics studio. Ingestion of 69 mg of clay would be required to result in an absorbed dose equal to the average dermal dose of 3.37 pg TEQ/d (this assumes the clay has an average concentration of 162 pg TEQ/g and 30% of the dioxin is absorbed during ingestion).

10. CONCLUSIONS

Hypothetical dioxin dose estimates were calculated for each subject assuming that all used a 20% ball clay blend with 162 pg TEQ/g. The single-day total doses across the 10 subjects ranged from 0.49 to 20.81 pg TEQ/d, with an average of 3.45 pg TEQ/d. The dermal dose was the major contributor to total dose, exceeding 78% for all subjects. Ingestion and inhalation contributed similar amounts, generally in the range of 1 to 10% of total dose. Hand and arm exposure accounted for much of the dermal dose for all subjects. The two subjects who wore summer clothing had foot and leg exposures accounting for about 44 to 79% of the dermal dose. Facial exposures were low accounting for less than 3% of total dermal dose.

Clay exposure was found to be highly dependent on the type of work being performed. Throwing clay on the wheel resulted in much higher clay exposures than did any other clay activities. This is due to the increased contact with clay while working on the wheel and the wet, sticky consistency of the clay needed for that work. Emptying bags and mixing dried clays also led to high exposures.

A Monte Carlo simulation was performed to model how doses could vary in a broad population of artists with exposures outside the hypothetical scenario evaluated in this study. The simulation, using a variety of assumed input distributions, suggests that doses could extend to levels higher or lower than those estimated for the hypothetical scenario. Also, it indicated that clothing, the fraction of ball clay in the blend and dioxin concentration contributed most to variance in total dose. Many of the input distributions used in this simulation were based on very limited data or judgment. Therefore, the simulation results are best interpreted as preliminary indications of how to extrapolate the observations of this study to a broader population, and further study is recommended to confirm these predictions.

In the general population, adult daily intakes of CDD/CDFs and dioxin-like polychlorinated biphenyls (PCBs) are estimated to average 43 and 23 pg TEQ, respectively, for a total intake of 66 pg TEQ/day (U.S. EPA, 2003). More than 90% of this intake is derived from food ingestion. These intake values are based on the “administered” dose or the amount taken into the body before absorption. The hypothetical doses presented in this report are on an absorbed dose basis. Thus, the background dose must be converted to an absorbed basis to compare it to the values presented here. U.S. EPA (2003) reports that about 80% of dioxins in foods are absorbed into the body. Applying this factor, the background dose on an absorbed basis is 34.4 and 18.4 pg TEQ/day for CDD/CDFs and dioxin-like PCBs, respectively, for a total intake of 52.8 pg TEQ/day. Comparing these values to the average of the hypothetical doses for the 10 subjects estimated here (3.45 pg TEQ/day) indicates that the ball clay dose is 10% of the background CDD/CDF dose and about 7% of the total CDD/CDF/PCB dose (on a TEQ basis). Note that the general population dioxin dose is a long-term average and the hypothetical ball clay dioxin dose is an estimate for a single day when exposure occurs. Accordingly, this comparison implies that ball clay use is a frequent event, so that the long-term daily average ball clay dose is similar to the single-day dose. If ball clay use is infrequent, then the long-term average dose from ball clay will be reduced and adjustments would be needed to make a valid comparison to the background dioxin dose.

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