Appendix G

Evaluation of Clay Dust Inhalation

APPENDIX G. EVALUATION OF CLAY DUST INHALATION

The methodology used to evaluate the dose of clay dust and associated dioxin received via inhalation is discussed in this appendix. The appendix is divided into four sections: clay dust size distribution, particle inhalability, respiratory deposition of clay dust, and delivered dose estimates.

CLAY DUST SIZE DISTRIBUTION

As discussed in the main body of this report, the size distribution of clay dust was measured using a Delron cascade impactor and a Climet during regular daily activities in the art studio. The Climet optically determines particle concentration for six size bins with the associated physical particle diameter (d_p) of 0.3–0.5, 0.5–1, 1–2.5, 2.5–5, 5–10, and >10 µm. Aerodynamic particle diameter (d_{ae}) can be estimated for the Climet's size bins by assuming that the airborne clay dust has a density of 2.6 g/cm^3, similar to that of bulk clay.[1] Using this approach, a clay particle with a d_p of 10 µm has a d_{ae} of 16 µm. The Delron cascade impactor fractionates particles directly, based on their d_{ae}, into the seven ranges of <0.5, 0.5–2, 2–4, 4–8, 8–16, 16–32, and >32 µm.

During normal artisan activities (Subjects 1–8), 64 ± 9% (mean ± SD) of the aerosol is associated with particles having a d_{ae} > 16 µm based on average Climet data. Based on average impactor data, 63 ± 13% of the aerosol is associated with a d_{ae} > 16 µm (Subjects 1–8). The particle size distributions to which the artisans were exposed was assumed to be log-normally distributed.[2] The cascade impactor data were selected for estimating particle size distributions for the following reasons: (1) the impactor measures particle size based on the aerodynamic behavior of particles, whereas the Climet uses light scattering to estimate a physical particle size; (2) the impactor affords a better characterization of the large particles than does the Climet because it contains an additional size bin of 16–32 µm; and (3) particle deposition in the respiratory tract is a function of d_{ae}. Thus, uncertainty in estimates of respiratory deposition is reduced by the direct measurement of d_{ae} by the impactor. The clay dust size distribution was not estimated for runs where two or more of the impactor stages were below the nondetect level.

When engaged in normal artisan activities, the mass median aerodynamic diameter (MMAD) of clay dust to which artisans were exposed ranged from 13 to 45 µm. Table G-1

[1] $d_{ae} = d_p \{(\text{clay density} * Cc(d_p)) / (H_2O \text{ density} * Cc(d_{ae}))\}^{0.5}$, where: $Cc(d_p)$ and $Cc(d_{ae})$ are the Cunningham slip correction factor for the physical and aerodynamic particle size, respectively. For more information, the reader is referred to ICRP (1994), page 239.

[2] For more information about particle sizing and the log-normal distribution, the reader is referred to Hinds (1999).

provides a characterization of clay dust exposures for each subject. Figure G-1 illustrates a log-probability plot of a typical (i.e., near the average MMAD) clay dust particle size distribution and a background sample from the studio. The prevalence of fewer large particles in the background aerosol can be explained easily, based on particle-settling velocities. The settling velocities for the d_{ae} of 1-, 10-, and 20-µm particles are 3.5×10^{-3}, 0.3, and 1.2 cm/s, respectively. Due to their rapidly settling velocities, large particles (d_{ae} > 10 µm) are maintained in the air only by active generation or resuspension from surfaces. The substantive presence of large particles (52% of mass associated with a d_{ae} > 10 µm) in the background sample is suggestive of particle resuspension due to movement (e.g., walking and setting up sampling equipment in the studio).

Table G-1. Clay dust size distribution and concentration during normal activities

Subject	Size distribution[a]		Total concentration (mg/m^3)
	MMAD (µm)	σ_g	
1	26.9	3.9	0.35
2	44.6	4.8	0.47
3	18.5	4.3	0.99
4	n.a.	n.a.	0.37
5	n.a.	n.a.	0.13
6	20.2	3.0	0.61
7	13.0	3.6	0.51
8	26.7	3.3	0.64
Mean ± SD	25.0 ± 11	3.8 ± 0.7	0.51 ± 0.25

[a]The aerosol size distribution is described in terms of the mass median aerodynamic diameter (MMAD) and geometric standard deviation (σ_g).
n.a. = not available

Data were also available for two subjects during specific activities (i.e., when sculpting and using a pottery wheel) (see Table G-2). During pottery wheel operations, an average MMAD of 33 µm with a geometric standard deviation (σ_g) of 5.4 was observed. A dog was present during two of the sculpting runs. The MMAD with the dog present was 21 µm versus only 16 µm without the dog. The shift toward larger particles when the dog was present appears to be consistent with particle resuspension due to the dog's movement around the studio.

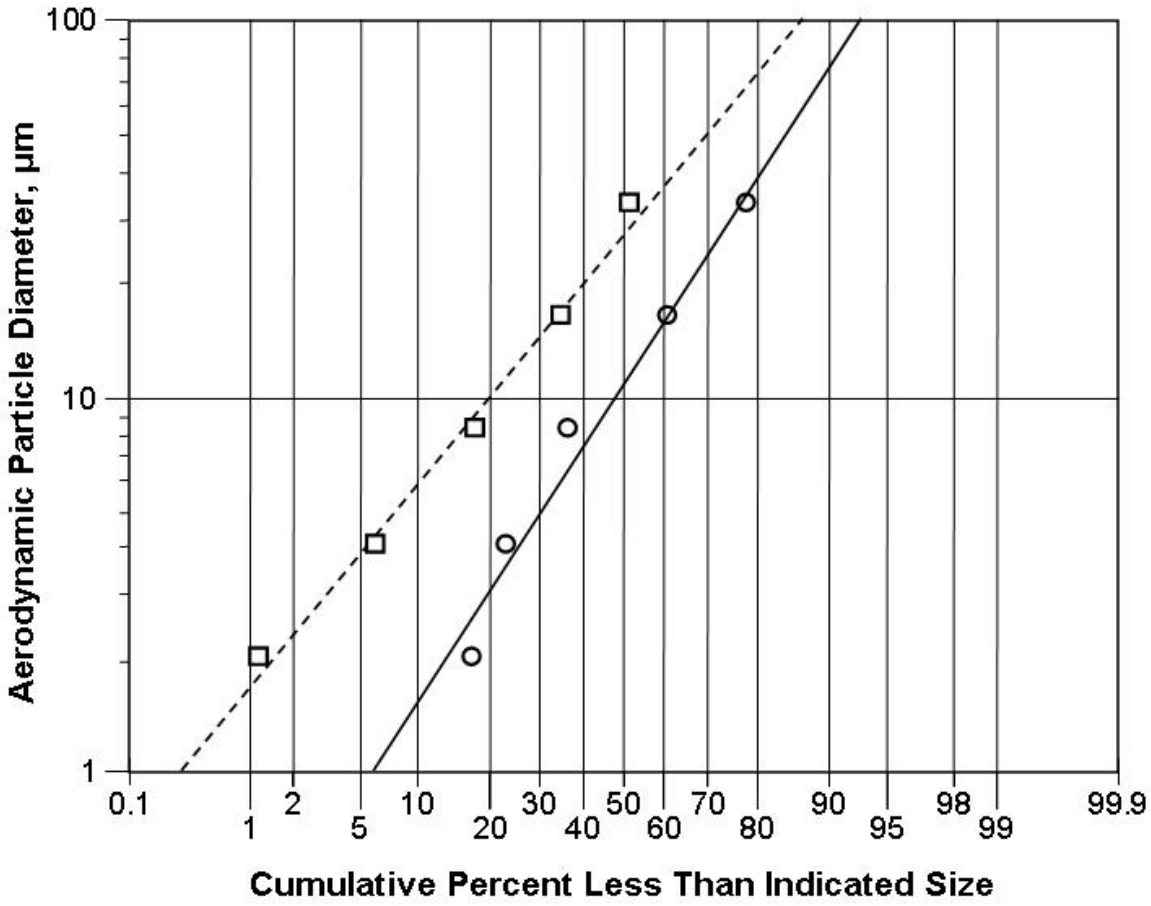

Figure G-1. Clay dust particle size distribution during normal artisan activities from analysis of cascade impactor data. Illustrated are the data for Subject 8 ( ) and a background sample when work was not being done in the studio (○). The dashed and solid lines illustrate the log-normal distribution for these respective data. The mass median aerodynamic diameter (MMAD) of clay dust was 27 µm (σ_g = 3.3) for Subject 8, whereas the background sample had an MMAD of 11 µm (σ_g = 4.6).

PARTICLE INHALABILITY

For a given particle size, inhalability is the ratio of the particle concentration that enters the respiratory tract through the nose or mouth to the concentration of these particles in the ambient air. Inhalability depends mainly on particle size (i.e., d_{ae}), route of breathing, wind speed, and a person's orientation with respect to wind direction. Wind speeds in the art studio were assumed to be 0.3 m/s or less (Baldwin and Maynard, 1998). The artisans were presumed to move about the studio such that their orientation was random with respect to wind direction.

Table G-2. Clay dust size distribution and concentration during specific activities

Subject		Size distribution[a]		Total concentration (mg/m^3)
		MMAD µm	σ_g	
Subject 9 (Pottery wheel)	Run 1	33.7	6.2	0.049
	Run 2	n.a.	n.a.	0.046
	Run 3	24.8	4.3	0.102
	Run 4	n.a.	n.a.	0.073
	Run 5	39.3	5.6	0.152
	Mean ± SD	32.6 ± 7.3	5.4 ± 0.9	0.085 ± 0.044
Subject 10[b] (Sculpting work)	Run 1	21.2	3.9	0.48
	Run 2	20.4	3.2	0.24
	Run 3	16.0	3.5	0.24

[a]The aerosol size distribution is described in terms of the mass median aerodynamic diameter (MMAD) and geometric standard deviation (σ_g).
[b]A dog was present during Runs 1 and 2 but not during Run 3. Therefore, these three runs were not averaged as was done in the case of the pottery wheel work.
n.a. = not available

The clay dust aerosol present under normal activities in the art studio was observed to have an average MMAD of 25 µm and σ_g of 3.8. Hence, 50% (on average, by mass) of the airborne clay dust is composed of particles having a d_{ae} of $\geq$25 µm, a size that is generally considered to be unable to penetrate the thorax (ACGIH, 2004). These large particles ($d_{ae} \geq 25$ µm), if inhaled, will deposit almost completely and exclusively in the extrathoracic (ET) airways. Thus, determining inhalability is key to estimating the delivered dose of these large particles. For smaller particles, inhalability still describes the fraction of airborne particles that may enter the respiratory tract and thereby the availability of these particles for deposition in the lung.

Only limited data are available on the inhalability of particles from calm air (wind speeds of 0.3 m/s and less). Inhalability from calm air depends on the route of breathing. Logistic functions describing particle inhalability during nasal [$P(I_N)$] and oral [$P(I_O)$] breathing are given by Ménache et al. (1995) and Brown (2005):

$$P(I_N) = 1 - \frac{1}{1 + \exp(10.32 - 3.1141n(d_{ae}))} \quad \text{(G-1)}$$

$$P(I_O) = \frac{1.44}{1 + 0.44\exp(0.0195 d_{ae})} \quad \text{(G-2)}$$

Note that these equations depend only on aerodynamic particle diameter, d_{ae}. Given by Eq G-1, $P(I_N)$ begins a rapid decline from 0.95 at d_{ae} = 11 µm, to 0.5 at d_{ae} = 27.5 µm, and 0.1 at d_{ae} = 56 µm. Equation G-2 predicts a slow decline in $P(I_O)$ from 0.95 at d_{ae} = 8 µm, to 0.5 at d_{ae} = 74 µm, and 0.1 at d_{ae} = 175 µm.

Figure G-2 illustrates particle inhalability predicted by Eqs G-1 and G-2 (shown by solid lines) along with relevant experimental data. Based on high wind speeds (1–8 m/s), the American Conference of Governmental Industrial Hygienists (ACGIH) inhalability criterion is also illustrated (shown by dashed lines) for comparative purposes. Equation G-1 for $P(I_N)$ describes the experimental nasal inhalability data well with an r^2 of 0.86 (model sum of squares divided by the total corrected sum of squares). A negative r^2 is obtained for the fit of the ACGIH (2004) criterion to these data.[3] Equation G-2 describes the experimental oral inhalability data with an r^2 of 0.69, whereas the ACGIH criterion fit with an r^2 of 0.32.

RESPIRATORY DEPOSITION OF CLAY DUST

Inhaled particles may be either exhaled or deposited in the ET, tracheobronchial (TB), or pulmonary (PU) airways. The deposition of particles in the respiratory tract depends primarily on inhaled particle size (i.e., d_{ae}), route of breathing (through the nose or mouth), tidal volume (V_T), and breathing frequency (f). Reference respiratory values for males and females were adopted from the International Commission on Radiological Protection (ICRP, 1994). In addition to breathing patterns (Table G-3) necessary for deposition calculations, males and females were assumed to have a functional residual capacity of 3,300 mL and 2,680 mL, respectively. The majority (70%) of the subjects were female; only Subjects 1, 2, and 5 were male.

Particle deposition in the respiratory tract was predicted using the publicly available Multiple Path Particle Dosimetry (MPPD) model.[4] The MPPD model was developed by the CIIT

[3]An r^2 is calculated as the model sum of squares (MSS) divided by the total corrected sum of squares (TSS). The MSS equals the TSS minus the residual sum of squares (RSS). In typical linear regressions, when a model is fitted to a data set, the resulting r^2 must be non-negative because the least square fitting procedure assures RSS $\leq$ TSS. When r^2 is computed on excluded data, i.e., data not used to fit the model, the RSS can exceed the TSS. In this case, r^2 (which is not the square of r) can be negative, indicating that the mean of the data is a better predictor than the model.

[4] The MPPD program is available on request from the CIIT Centers for Health Research (<asgharian@ciit.org>).

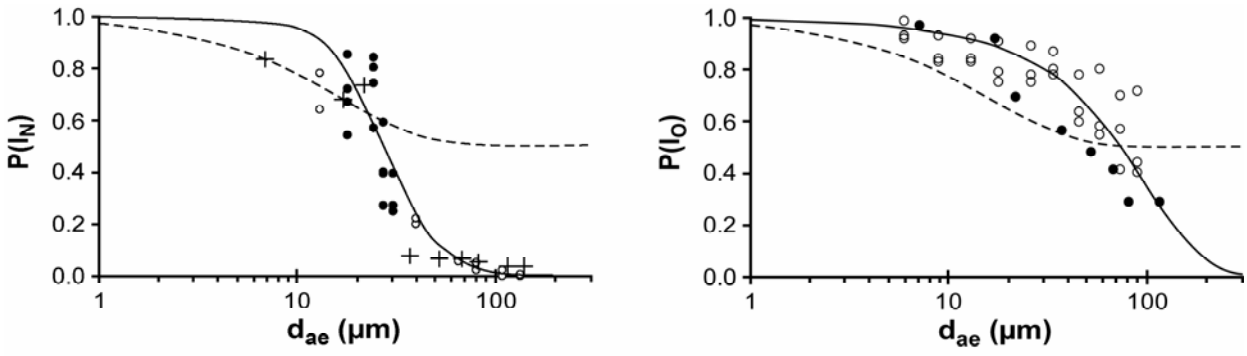

Figure G-2. Particle inhalability from calm air for nasal [$P(I_N)$] and oral [$P(I_O)$] breathing as a function of aerodynamic particle diameter (d_{ae}). Left panel [── Equation G-1, ● Breysse and Swift (1990), + Hinds et al. (1998), ○ Hsu and Swift (1999), - - - ACGIH (2004)]. Right panel [── Equation G-2, ○ Aitken et al. (1999), ● Kennedy and Hinds (2002), - - - ACGIH (2004)].

Table G-3. Breathing patterns used in particle deposition calculations[a]

Activity		Males	Females
Sitting	V_T (mL)	750	464
	f (min^{-1})	12	14
Light exercise	V_T (mL)	1,250	992
	f (min^{-1})	20	21

Source: ICRP (1994), Table 8.

1

2

Centers for Health Research (CIIT), United States, in collaboration with the National Institute of Public Health and the Environment (RIVM), the Netherlands, and the Ministry of Housing, Spatial Planning and the Environment, the Netherlands. The MPPD model may be used to predict the deposition in the human respiratory tract for particles between 0.01 and 20 µm in diameter. In the lung, the model considers deposition by the mechanisms of impaction, sedimentation, and diffusion. Additional model details are available elsewhere (DeWinter-Sorkina and Cassee, 2002). For the size of the clay dust, only impaction and sedimentation are of concern.

3 Using the MPPD model, deposition was predicted for the ET, TB, and PU regions of the
4 respiratory tract. Particle deposition was estimated individually for oral and nasal breathing.
5 During oral breathing, deposition in the TB airways did not always reach zero by a d_{ae} of 20 µm

(the upper limit for the MPPD model). For d_{ae} > 20 µm, deposition in the TB airways was estimated by a best fit polynomial (3rd or 4th degree) determined using CurveExpert 1.3 (112B Crossgate St., Starkville, MS 39759). This polynomial function was fitted to TB deposition fractions for d_{ae} from 10 to 20 µm. The predicted ET deposition during oral breathing for a d_{ae} >20 µm was taken as one minus the TB deposition fraction for oral breathing. For nasal breathing, these additional steps were unnecessary because TB deposition was well under 1% at a d_{ae} of 20 µm.

External to the MPPD model, all of the predicted deposition fractions were corrected for particle inhalability using Eqs G-1 and G-2. The current version of MPPD model offers an inhalability correction for nasal breathing only. For a given d_{ae}, an inhalability corrected deposition fraction is the product of the uncorrected deposition fraction and the predicted inhalability for that d_{ae}. Unless otherwise specified, all mention of particle deposition fractions in the main body of this report and subsequently in this appendix refer explicitly to inhalability corrected deposition fractions.

The deposition fraction (DF_r) of an aerosol in a region of the respiratory tract is the integral of the deposition fractions across all particle sizes in the aerosol:

$$DF_r(MMAD, \sigma_g) = \int_0^{\infty} DF_r(d_i)\rho(d_i)\delta d_i \qquad \text{(G-3)}$$

where:

$DF_r(d_i)$ = the deposition fraction in region, r, of particles having an aerodynamic diameter of d_i

$\rho(d_i)$ = the mass fraction associated with the interval δd_i

The total deposition fraction for the respiratory tract is the sum of DF_r for the ET, TB, and PU regions. Equation G-3 can be approximated by summing the particle deposition fractions at known intervals or percentiles of the particle size distribution. Here, the interval of 1% was used and the approximation is:

$$DF_r(MMAD, \sigma_g) \approx \frac{1}{100} \sum_{P=0.01}^{0.99} DF_r(d_i) \qquad \text{(G-4)}$$

where:

$DF_r(d_i)$ = the deposition fraction in region, r, of particles having an aerodynamic diameter d_i (the particle size associated with a given percentile, P, of the size distribution).

For a log-normal distribution, d_i is given by:

$$d_i = MMAD\ \sigma_g^{z(P)} \qquad \text{(G-5)}$$

where:

z(P) = the normal standard deviate for a given probability

Table G-4 provides the predicted regional deposition fractions for the clay dust in the respiratory tract of each subject for oral and nasal breathing at two activity levels. These deposition fraction estimates were based on each subject's measured aerosol exposure size distribution (see Tables G-1 and G-2). Subjects 4 and 5 lacked aerosol size distribution data and were assumed exposed to an aerosol with an MMAD of 25 μm and σ_g of 3.8, this being the average for artisans during normal activities (see Table G-1). The deposition fraction estimates for Subject 10 were based on Run 3, when the dog was not present in the studio.

DELIVERED DOSE ESTIMATES

The rate of particle deposition in a region of the respiratory tract may be expressed as:

$$\dot{D}_r(t) = C(t)\quad (t)V_T(t)DF_r(t) \qquad \text{(G-6)}$$

where:

$\dot{D}_r$ = the rate of deposition per unit time in region r
C = the exposure concentration
f = breathing frequency
V_T = tidal volume
DF_r = the deposition fraction in region r

Note that all of the variables in Eq G-6 may vary with time. The dose to a respiratory region is determined by integrating Eq G-6 over the exposure duration.

Table G-4. Regional deposition fractions (corrected for inhalability) for clay dust in the respiratory tract

	Sitting						Light exercise					
	Nasal breathing			Oral breathing			Nasal breathing			Oral breathing		
Subject	ET	TB	PU	ET	TB	PU	ET	TB	PU	ET	TB	PU
1	0.441	0.015	0.022	0.473	0.082	0.058	0.473	0.006	0.011	0.516	0.060	0.052
2	0.336	0.011	0.016	0.412	0.059	0.042	0.360	0.004	0.008	0.442	0.044	0.037
3	0.472	0.028	0.033	0.431	0.104	0.067	0.531	0.010	0.020	0.486	0.074	0.075
4	0.447	0.021	0.022	0.471	0.091	0.050	0.487	0.007	0.013	0.521	0.064	0.056
5	0.458	0.016	0.023	0.479	0.086	0.061	0.492	0.006	0.011	0.523	0.063	0.054
6	0.526	0.023	0.022	0.521	0.108	0.053	0.566	0.007	0.012	0.581	0.075	0.059
7	0.549	0.035	0.041	0.432	0.128	0.085	0.622	0.013	0.025	0.498	0.090	0.095
8	0.451	0.018	0.017	0.507	0.087	0.041	0.483	0.005	0.010	0.557	0.061	0.046
9	0.368	0.020	0.023	0.396	0.077	0.047	0.410	0.007	0.014	0.437	0.054	0.053
10	0.533	0.030	0.033	0.462	0.118	0.072	0.593	0.010	0.020	0.525	0.083	0.081

ET = extrathoracic; PU = pulmonary; TB = tracheobronchial

By assuming that aerosol characteristics and an individual's activity levels are fairly constant over discrete periods of time, the dose to a respiratory region may be approximated by:

$$D_r = 0.06\sum_{j=1}^{n}(V_T f)_j (CT)_j [F_m DF_{m,r} = F_N DF_{N,r}]_j \qquad \text{(G-7)}$$

where:

D_r = the dose (μg) to region r of the respiratory tract
V_T and f = tidal volume (mL) and breathing frequency (min^{-1}) for a specified activity j
C and T = exposure concentration (mg/m^3) and duration (hr) during activity j
F_m and F_N = the fraction of a breath entering the respiratory tract through the mouth and nose, respectively, during activity j
$DF_{m,r}$ and $DF_{N,r}$ = the deposition fraction for oral and nasal breathing, respectively, in region r of the respiratory tract while performing activity j
Constant 0.06 = a unit conversion parameter

As expressed, an "activity" in Eq G-7 could be associated with changes in exposure concentration, the particle size distribution, and/or an individual's exertion level. For simplicity, only two exertion levels (sitting and light exercise) and a single particle size distribution (see Tables G-1 and G-2) were considered for each subject.

The fraction of flow through the mouth (F_m in Eq G-7) increases with activity level and varies between individuals. For the two activity levels considered here, most people (87%) will breathe through their nose (Niinimaa et al., 1981). Hence, for these people, F_m= 0 and F_N= 1 in Eq G-7. However, 13% of people will be oronasal breathers even at rest, i.e., they will breathe simultaneously through the nose and mouth (Niinimaa et al., 1981). This latter group is commonly referred to in the literature as "mouth breathers" (e.g., ICRP, 1994). Derived from Niinimaa et al. (1981), the fraction of air respired through the mouth (F_m) is well described by a modified exponential function in the form of:

$$F_m = \alpha \exp\left(\frac{\gamma}{\dot{V}_e}\right) \qquad \text{(G-8)}$$

where:

$\dot{V}_e$ = minute ventilation

α = 0.748 and γ=-7.09 (r^2=0.997) in mouth breathers for 10≤$\dot{V}_e$≤80 L/min and

35.3≤$\dot{V}_e$≤80 L/min, α = 0.744, and γ=-18.3 (r^2=0.998) in normal augmenters

For $\dot{V}_e$ <35.3 L/min, normal augmenters breathe entirely through the nose, i.e., F_m = 0. F_N is one minus F_m regardless of the activity.

Table G-5 gives the estimated clay dust doses to regions of the respiratory tract for each subject during nasal and oronasal breathing. Estimates are for a 4-hour exposure assuming that the exposed individual spent 50% of his or her time sitting and 50% engaged in light exercise. For oronasal breathing in Table G-5, there is a small positive bias in ET doses and a corresponding negative bias in TB doses calculated by Eq G-7. In other words, this method of calculating ET and TB doses shifts the pattern of deposition toward the head relative to the real-life pattern of deposition. This shift occurs due to deposition being calculated at a higher airflow rate through the nose and mouth than actually occurs during oronasal breathing. The deposition calculations presumed that all inhaled airflow was through the nose or mouth. In reality, inhaled air is partitioned between the nose and the mouth, and the actual flows (for sitting and light exercise) are roughly half of that used in the deposition calculations. For breathing by a single

route (nasal or oral), changing activity from sitting to light exercise approximately triples flow rates but only slightly increases ET deposition and modestly decreases TB deposition (see Table G-4). The effect of using Eq G-7 for calculating doses during oronasal breathing should similarly affect the pattern of deposition. Ultimately, particles deposited in the ET and TB regions will typically be cleared to the throat and swallowed within 24 to 48 hours postdeposition (ICRP, 1994). Hence, the exact site of deposition (i.e., ET versus TB) is of little significance because both regions effectively contribute to ingested doses.

Table G-6 provides estimates of the dioxin absorption in each subject for nasal and oronasal breathing. Particles deposited in the ET and TB regions clear rapidly (within 1–2 days) to the throat and are swallowed. The absorption of dioxin from particles deposited within the ET and TB regions was treated as if the particles had been ingested. Dose estimates for oronasal breathing are slightly more conservative from a safety or risk perspective than presuming nasal breathing. However, nasal breathing may be considered as representative of the majority of the population (87%). Oronasal breathing is thought to represent 13% of healthy individuals (Niinimaa et al., 1981). In contrast to healthy subjects, Chadha et al. (1987) found that the majority (11 of 12) of patients with asthma or allergic rhinitis breathe oronasally even at rest. On average across all the subjects, dioxin doses are about 1.2 times greater for oronasal than for nasal breathing.

Table G-5. Regional doses (μg) of clay dust in the respiratory tract[a]

	Nasal breathing			Oronasal breathing		
Subject	**ET**	**TB**	**PU**	**ET**	**TB**	**PU**
1	664	12	20	693	53	48
2	678	11	19	757	52	47
3	1,677	47	75	1,612	143	154
4	580	13	19	598	45	41
5	256	4.6	7.7	264	21	19
6	1,114	22	29	1,126	85	70
7	1,011	30	49	917	90	100
8	997	18	24	1,067	72	57
9	110	2.9	4.5	114	8.8	9.2
10	455	12	18	431	39	39
Mean	754	17	27	758	61	58
SD	460	13	21	445	39	42

[a] Doses calculated by Eq G-7 as described in the text.
ET = extrathoracic; PU = pulmonary; TB = tracheobronchial

Table G-6. Estimates of dioxin absorption[a] (pg TEQ)

Subject	Nasal breathing			Oronasal breathing		
	ET and TB[b]	PU[c]	Total	ET and TB[b]	PU[c]	Total
1	0.033	0.003	0.035	0.036	0.006	0.043
2	0.034	0.003	0.036	0.039	0.006	0.045
3	0.084	0.010	0.094	0.085	0.020	0.105
4	0.029	0.002	0.031	0.031	0.005	0.037
5	0.013	0.001	0.014	0.014	0.002	0.016
6	0.055	0.004	0.059	0.059	0.009	0.068
7	0.051	0.006	0.057	0.049	0.013	0.062
8	0.049	0.003	0.052	0.055	0.007	0.063
9	0.005	0.001	0.006	0.006	0.001	0.007
10	0.023	0.002	0.025	0.023	0.005	0.028
Mean	0.038	0.004	0.041	0.040	0.007	0.047
SD	0.023	0.003	0.026	0.023	0.006	0.029

[a] Dioxin concentration was assumed to be 162 pg toxic equivalent (TEQ) per gram clay.
[b] Absorption fraction of 0.3 assumed, extrathoracic (ET) and tracheobronchial (TB) rapidly clear into the gastrointestinal tract.
[c] Absorption fraction of 0.8 assumed, due to slow clearance from pulmonary (PU) region.

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