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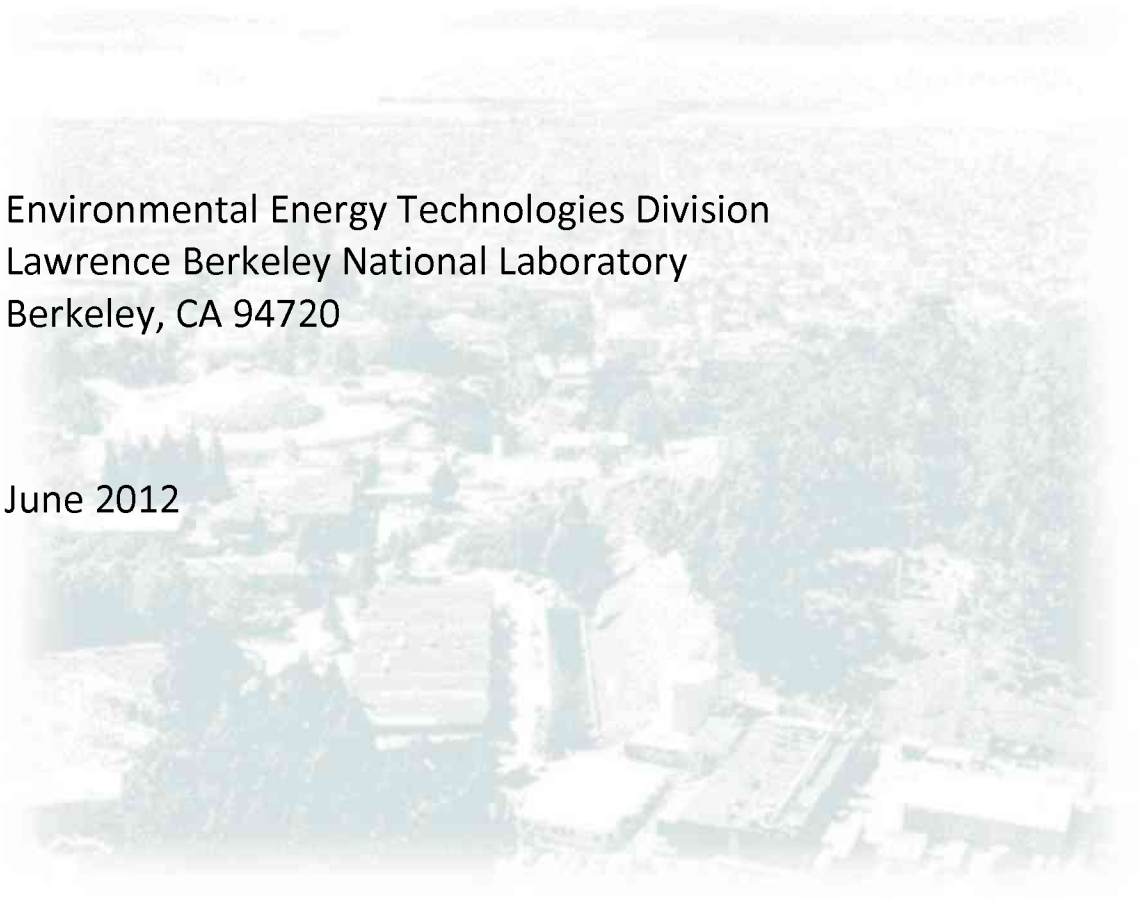

Computing Toxic Load for Shelter-in-Place Analysis Using Joint Urban 2003

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ABSTRACT

Toxic load is a metric used to quantify acute effects from inhalation exposures. The toxic load to individuals depends on the fluctuations of chemical concentrations because the exposure-response relationship is often nonlinear. To study the effects of concentration fluctuations on toxic load, we analyzed concentration data from the Joint Urban 2003 series of city-scale tracer gas studies. We summarized the outdoor concentrations measured by fast-response gas analyzers and computed outdoor toxic load. We then analyzed the amount of tracer gas that entered a 16-story office building and compared indoor and outdoor toxic loads. We show that the time aggregation used to compute toxic load is a source of uncertainty in hazard assessments. However, in comparison to other uncertainties and variability inherent in hazard assessments involving buildings, the choice of time averaging interval plays a minor role, especially when toxic load is used to compute other aggregated metrics such as the effectiveness of sheltering indoors. In the absence of compound-specific information to dictate a time average, we propose following Acute Exposure Guideline Levels (AEGLs) and using timescales on the order of minutes for computing indoor toxic load.

KEYWORDS: Acute health effects; nonlinear dose response; indoor-outdoor exposure; toxic releases; hazard assessment

1. INTRODUCTION

Computing the indoor effects from an outdoor toxic release requires predicting the outdoor dispersion of the chemical, the infiltration of the chemical into the building, the mixing of the chemical with indoor air, and the acute effects from the exposure. Recent studies have modeled the transport and dispersion of chemical compounds both outdoors and indoors under various release scenarios [1-4]. Model predictions of outdoor and indoor concentrations vary with time, but there is no consensus on how to relate time-varying air concentrations to health effects [5-8]. Moreover, exposure guidelines and limits are typically established assuming exposure to a constant concentration. For example, Acute Exposure Guideline Levels (AEGLs) [9], which are commonly used by emergency responders to assess health risks from toxic releases, set threshold exposure limits for the general public that range from ten minutes to eight hours. Most of the toxicological data used to develop AEGLs are based on experiments conducted at constant concentrations.

However, concentration fluctuations may be important for computing the acute health effects from inhalation of toxic chemicals because the exposure-response relationship is often nonlinear. Ten Berge et al. [10] used a probit analysis to estimate the mortality response to irritant chemicals and systemically acting chemicals for different species. Their analysis suggests that toxic load $TL = C^n T$ is a predictor for mortality response, where C and T are the concentration and time of exposure, and n is the toxic load exponent that is often different from 1. For many chemicals, an n between 0.8 and 3.5 is suggested for computing acute inhalation toxicity.

The toxicological studies reviewed by ten Berge et al. [10] were mostly animal tests in which the chemical concentration was held steady throughout the experiment. It is thus unclear

to what degree the derived value of n applies to exposures to highly fluctuating chemical concentrations. The absence of toxicological data highlights important questions for current hazard assessments: How does one compute a reasonable and representative toxic load from time-resolved concentrations, given the absence of detailed compound-specific experiments? And how does one then compare computed toxic load to health effects, which are also established under laboratory conditions of a constant chemical concentration?

To address these questions, we examined data that, in part, we collected during Joint Urban 2003 (JU2003). JU2003 is a series of city-scale field experiments conducted in the downtown area of Oklahoma City [11]. Its research goal was to study the dispersion of pollutants in urban areas, specifically in the presence of tall buildings that may cause complex airflows in street canyons. Ten sets of tracer gas experiments were conducted to characterize these urban effects and any potential day-versus-night differences on plume dispersion. In this paper, we use the JU2003 dataset to estimate the difference in exposure and acute effects if people were to take shelter in the building as opposed to remaining outdoors. We ask the question: To what degree are the computed toxic load and shelter-in-place (SIP) effectiveness affected by the choice of time averaging interval of the concentration data?

This paper focuses on the JU2003 data from a 16-story office building where indoor tracer gas measurements were made during the outdoor gas experiments. We use the inert tracer gas data to illustrate how the resulting toxic load is influenced by the time aggregation of the concentration data. Detailed description of the various experiments and equipment used is provided by Allwine and Flaherty [11] and Black et al. [12].

2. TOXIC LOAD AND SHELTER-IN-PLACE EFFECTIVENESS

Our prior work on evaluating SIP effectiveness during large-scale chemical releases [2-4] assumed that the toxic load applies to time-varying concentrations in the following form:

$$TL = \int_0^T [C(t)]^n \, dt \qquad (1)$$

If the exposure concentration, $C(t)$, is not constant but varies with time, and when n is not unity, the values of TL would differ when different levels of time aggregation are applied to the concentration data. In idealized cases [2, 13], Eq. (1) provides a simple description of the nonlinear exposure-response relationship by the exponent n. But in real atmospheric release conditions, airborne chemical concentrations can fluctuate greatly, as measured using fast-response instruments [14]. For large values of n, Eq. (1) might overemphasize the importance of very short and intermittent concentrations on health effects, when data or experience suggests otherwise. This raises the question of whether TL computed from highly time-resolved concentration data using Eq. (1) is suitable for comparison with acute inhalation toxicological data in assessing health effects. Because toxicity experiments are conducted in the laboratory at constant air concentrations, and typically for longer durations, some aggregation of time-varying concentrations is needed in order to be consistent with the toxicological data.

When $n > 1$, which is commonly the case for acutely toxic compounds [10], the value of TL decreases with increasing time aggregation. TL converges to a lower limit that is defined by $\overline{C}^n T$ where $\overline{C}$ is the time-averaged exposure concentration and T is the exposure time. Following this reasoning, Ride [8] introduced the notion of a toxic load ratio, TLR, to relate the toxic load of fluctuating concentrations to that of constant concentrations with the same mean value:

$$TLR = \frac{\int_0^T [C(t)]^n \, dt}{\overline{C}^n T} \qquad (2)$$

Ride [8] computed the value of *TLR* for two experiments that collected concentration data at frequent time intervals (20 and 100 Hz) at close proximity to the release (20 and 140 m from the source). He suggested that the finest timescale suitable for estimating toxic load is the duration of a human breath, which is on the order of a few seconds. *TLR* is a useful measure also because when $n = 2$ or 3, *TLR* can be expressed in terms of two parameters, the coefficient of variation and skewness, that describe concentration fluctuations [8].

In our analysis, we used Eq. (2) and the outdoor and indoor tracer gas concentrations from JU2003 to compute $TLR_{n,out}$ and $TLR_{n,in}$. We computed these two terms for different levels of time averaging interval, and for n between 1 and 3, which is the range used in developing AEGLs [15]. In the absence of chemical-specific data to determine the value of n, AEGLs are developed using an assumed exponent of either 1 or 3. When toxicity data are available only for long exposures (e.g., hours), AEGLs are derived assuming an exponent of 3 in order to extrapolate the exposure limits to shorter time periods (e.g., minutes). Alternatively, when extrapolating from shorter-duration exposure to longer-duration exposures, an exponent of 1 is assumed. In either case, the value of n is selected to avoid underestimating the exposure limits caused by the extrapolation.

Many methods exist to compute the effectiveness of sheltering in place, such as comparing the expected health outcomes, or exposures, if a population were indoors versus outdoors during a release. In this paper, we define SIP effectiveness as the ratio of the indoor to the outdoor toxic load, *IO* (Eq. (3a)). *IO* describes the difference in expected adverse human outcomes for an individual standing indoors instead of just outside the building. Since TL_{out} and

TL_{in} are both quantities that depend on the level of time aggregation used in their computations (Eq. (1)), so too does IO.

We also define a term, Φ, as the ratio of the indoor and outdoor toxic load ratios (Eq. (3b)) to show how levels of time aggregation impact IO. For example, Φ approaches one when the effectiveness of SIP is insensitive to time aggregation, and Φ approaches zero when the effectiveness of SIP may be underestimated due to time aggregation in computed TL.

$$IO = \frac{TL_{in}}{TL_{out}} \tag{3a}$$

$$\Phi = \frac{IO\Big|_{\int_0^T [C(t)]^n dt}}{IO\Big|_{\bar{C}^n T}} = \frac{TLR_{in}}{TLR_{out}} \tag{3b}$$

3. JOINT URBAN 2003 DATASET

3.1. Tracer Gas Measurements

Finn et al. [16] analyzed the tracer gas concentrations measured by fast-response analyzers, which are summarized in Table 1. Of the various JU2003 experiments, referred to as Intensive Operating Periods (IOPs), we considered a subset where tracer gases were measured indoors and outdoors simultaneously: IOPs 4–10.

Outdoors, the mean sulfur hexafluoride (SF_6) concentrations were 2–10 ppb during daytime releases (IOPs 4–6), and 4–50 ppb during nighttime releases (IOPs 7–10) (Table 1). In the analysis that follows, we examine data recorded at two outdoor samplers that were located closest to the building studied. We excluded cases when the mean outdoor concentrations fell below 1 ppb. For IOPs 4–8, the two closest outdoor samplers were fast-response SF_6 tracer gas analyzers (TGAs) located approximately 60 m from the study building (samplers I and II in

Table 1). The TGAs measured SF_6 every one second. For IOPs 9–10, the closest samplers were Miran analyzers located a few meters away from the study building (samplers III and IV in Table 1). The Mirans measured SF_6 every half second. In general, higher concentrations were measured during IOPs 9–10, partly because the building was located closer to the release location (45 m), compared to 325 m for IOPs 4–7, and 170 m for IOP 8.

Table 1. Summary statistics of outdoor tracer gas concentrations measured by fast-response gas analyzers: mean (μ), standard deviation (σ), coefficient of variation (I), and skewness (S).

IOP	Release Time (CDT)	μ (ppb)	σ (ppb)	I (-)	S (-)	μ (ppb)	σ (ppb)	I (-)	S (-)
		Sampler I				Sampler II			
4[a]	11:00–11:30	2.8	3.4	1.2	1.2	5.4	5.0	0.9	1.1
	13:00–13:30	5.5	4.6	0.8	-0.03	3.4	4.6	1.4	2.0
	15:00–15:30	3.4	3.9	1.2	0.8	3.6	4.2	1.2	0.7
5[a]	09:00–09:30	2.9	3.9	1.3	1.2	4.9	5.0	1.0	0.4
	11:00–11:30	0.2	0.3	1.5	1.7	10.3	7.5	0.7	-0.04
	13:00–13:30	3.2	3.0	0.9	0.8	0.08	0.1	1.6	3.0
6[a]	09:00–09:30	2.4	3.3	1.4	1.5	5.0	6.0	1.2	1.3
	11:00–11:30	4.5	3.6	0.8	0.4	1.6	2.8	1.7	3.7
	13:00–13:30	2.4	2.8	1.2	1.2	2.0	2.9	1.5	1.8
7[a]	23:00–23:30	4.9	2.6	0.5	-0.4	3.6	4.4	1.2	1.9
	01:00–01:30	0.2	0.5	1.9	3.2	11.4	7.8	0.7	-0.2
8[b]	23:00–23:30	4.4	2.4	0.6	-0.04	–	–	–	–
	01:00–01:30	4.1	2.6	0.6	-0.2	–	–	–	–
	03:00–03:30	4.8	3.2	0.7	-0.03	–	–	–	–
		Sampler III				Sampler IV			
9[c]	23:00–23:30	8.1	13.0	1.6	4.2	10.9	14.5	1.3	3.2
	01:00–01:30	17.2	29.4	1.7	4.5	20.5	30.8	1.5	3.9
	03:00–03:30	12.6	27.4	2.2	7.5	38.9	62.0	1.6	2.8
10[c]	21:00–21:30	6.4	15.7	2.5	5.0	17.9	14.4	0.8	1.0
	23:00–23:30	8.2	10.0	1.2	2.9	28.3	34.1	1.2	1.8
	01:00–01:30	27.6	43.5	1.6	1.9	46.6	49.3	1.1	1.0

Release locations with respect to the studied building: [a] 325 m SW, [b] 170 m S, and [c] 45 m SW.
Outdoor sampler locations with respect to the studied building where indoor concentrations were measured concurrently: (I) 50 m WSW, (II) 65 m SE, (III) 10 m NW, and (IV) 5 m N.

The subject building, Building A, consists of 16 floors, of which two are below grade. The total floor space is approximately 18,600 m^2 (200,000 ft^2). The building is occupied by multiple tenants and has a mix of open floorplan and individual offices. The lower floors (2–7) are served by a mechanical room located on the fifth floor. The upper floors (8–14) are served by

a separate mechanical room located on the rooftop. Each of the above-grade floors consists of two perimeter ventilation zones and two central zones. Outside air is drawn from the street-level grates for below-grade floors, and from the rooftop grilles for floors 2–14. The ceiling plenum (space created by a drop ceiling) serves as an unducted return to a central return shaft.

Lawrence Berkeley National Laboratory and the National Institute for Occupational Safety and Health conducted the indoor sampling at Building A. Samples were taken at four locations, every four minutes, with a gas chromatograph with an electron capture detector (Table 2). Black et al. [12] provide an overview of the indoor sampling methodologies and the equipment used.

Indoors, continuous SF_6 measurements were collected during the three 30-minute continuous releases that took place during each IOP (Table 2). The tracer gas concentrations measured at the air intakes were similar to the outdoor air samples shown in Table 1, where values were the lowest during the daytime releases (IOPs 4–6), somewhat higher during the first two sets of nighttime releases (IOPs 7–8), and highest during the last two sets of nighttime releases (IOPs 9–10). Concentrations measured inside the rooftop mechanical room were about 30% lower than at the air intakes. Concentrations measured inside the fifth floor mechanical room were about a factor of four lower than in the rooftop mechanical room. The HVAC system was operating at minimum outdoor air for IOPs 4 and 6, the typical summertime mode when the building is occupied. The tracer gas data suggest a decay rate of about 1.0 h^{-1} in the rooftop mechanical room and 0.2 h^{-1} in the fifth floor mechanical room (Table 3). The mechanical system was turned off during all other IOPs. Tracer gas concentrations over time were more variable when the mechanical system was turned off. As a result, the decay rates only reflected

the change in tracer gas concentrations locally, and may not reflect the condition of the whole building.

Table 2. Summary statistics of tracer gas measurements inside Building A: mean (μ) and standard deviation (σ) in units of ppb.

IOP	Time of Day (CDT)	Rooftop Air Intake–East		Rooftop Air Intake West		Rooftop Mechanical Room		Fifth Floor Mechanical Room	
		μ	σ	μ	σ	μ	σ	μ	σ
4 [a]	11:01–12:01	2.3	2.1	2.0	2.5	0.9	0.4	0.5	0.2
	13:01–14:01	2.2	2.4	2.5	3.3	1.3	0.4	0.9	0.2
	15:03–16:00	1.7	1.9	1.4	1.3	1.2	0.2	1.1	0.1
5 [b,c]	09:07–10:01	1.4	2.2	1.6	2.5	0.6	0.3	0.1	0.1
	11:05–12:03	1.0	1.5	0.3	0.5	0.8	0.7	0.2	0.1
	13:03–13:46	1.0	1.1	1.8	1.8	0.3	0.1	0.3	0.1
6 [a]	09:03–10:01	3.0	4.2	1.6	1.7	0.8	0.4	0.3	0.2
	11:00–12:01	1.8	2.3	2.4	3.0	1.2	0.5	0.7	0.2
	13:01–14:03	1.7	2.2	1.4	1.5	1.1	0.3	0.9	0.1
7 [b,c]	23:03–00:03	5.8	6.0	4.9	5.7	6.1	4.2	1.4	1.2
	01:04–02:01	1.1	1.3	0.8	1.5	1.5	1.2	1.3	0.1
8 [b]	23:01–00:03	6.6	7.0	6.9	8.2	6.4	3.2	0.5	0.5
	01:00–02:01	7.6	8.7	8.9	11.9	6.8	3.6	1.2	0.2
	03:01–04:02	7.9	9.2	8.2	11.9	7.3	3.3	7.4	4.1
9 [b]	23:00–00:03	16.4	17.9	28.9	30.6	18.1	14.2	1.2	1.1
	01:00–02:01	21.3	25.4	24.6	24.4	20.1	14.5	2.9	0.5
	03:01–04:01	23.7	24.8	25.0	30.0	13.8	7.9	3.9	0.2
10 [b]	21:04–22:01	39.5	45.6	25.0	30.8	27.5	23.5	7.3	5.2
	23:04–00:01	34.3	33.3	24.7	25.5	20.3	16.3	5.4	1.3
	01:06–02:02	8.7	8.0	1.4	0.8	1.3	0.8	5.9	0.8

[a] HVAC running at minimum outdoor air.
[b] HVAC turned off.
[c] Protective measures, e.g., use of plastic sheet and tape, were also taken in safe rooms on certain floors.

Table 3. Time constant, k (h^{-1}), that describes the first-order decay of tracer gas in Building A. Standard error of k resulting from linear regression is shown in parentheses.

IOP	Rooftop Mechanical Room			Fifth Floor Mechanical Room		
	Release #1	Release #2	Release #3	Release #1	Release #2	Release #3
4	0.8 (0.04)	0.9 (0.04)	0.8 (0.19)	0.2 (0.01)	0.2 (0.01)	0.2 (0.01)
5	1.9 (0.36)	2.9 (0.16)	0.5 (0.10)	0.4 (0.09)	0.7 (0.03)	0.4 (0.04)
6	0.9 (0.07)	0.9 (0.07)	1.1 (0.13)	0.2 (0.01)	0.2 (0.01)	0.2 (0.01)
7	5.4 (2.50)	5.3 (0.61)	0.5 (0.04)	0.8 (0.02)	0.3 (0.01)	0.1 (0.003)
8	2.6 (0.22)	1.2 (0.05)	0.6 (0.05)	0.5 (0.04)	0.1 (0.05)	0.2 (0.04)
9	2.8 (0.21)	1.5 (0.26)	1.0 (0.23)	0.4 (0.03)	0.1 (0.02)	0.2 (0.03)
10	4.1 (0.20)	2.9 (0.14)	2.8 (0.16)	1.0 (0.04)	0.4 (0.01)	0.1 (0.01)

3.2. Concentration Averaging

Values of $TLR_{n,out}$ were calculated from averages of the outdoor concentrations at 5-second, 1-minute, and 4-minute time intervals. Elevated outdoor concentrations were measured from 25 to 44 minutes at the face of the building following a release. Nighttime releases (IOPs 7–10) tended to linger a few minutes longer than the daytime releases (IOPs 4–6).

Values of IO were computed from the indoor and outdoor concentration data. We averaged the measurements at the east and west rooftop air intakes to represent the outdoor concentrations at the building envelope, and we averaged the concentrations measured inside the mechanical rooms to represent the indoor concentrations on upper and lower floors. We calculated Φ using the 4-minute averaged concentration data.

4. RESULTS

4.1. Effects of Time Aggregation on Toxic Load Estimates

We computed TLR using Eq. (2) with $n = 2$ and 3 for all experiments. Figure 1 shows that $TLR_{n=2,out}$ is typically less than 4 for daytime releases, and less than 6 for the nighttime releases. This means that when $n = 2$, the time aggregation of the concentration data would change the toxic load estimates by a factor of 6 at most. Greater differences occurred only once during IOP 9 and once during IOP 10, due to a few concentration peaks that were substantially greater than the other measurements. For $n = 3$, Figure 1 shows that the differences in toxic load from exposure to outdoor concentrations as a result of time aggregation are typically less than 30 for $n = 3$. This range excludes the two extreme values that occurred once each during IOPs 9 and 10.

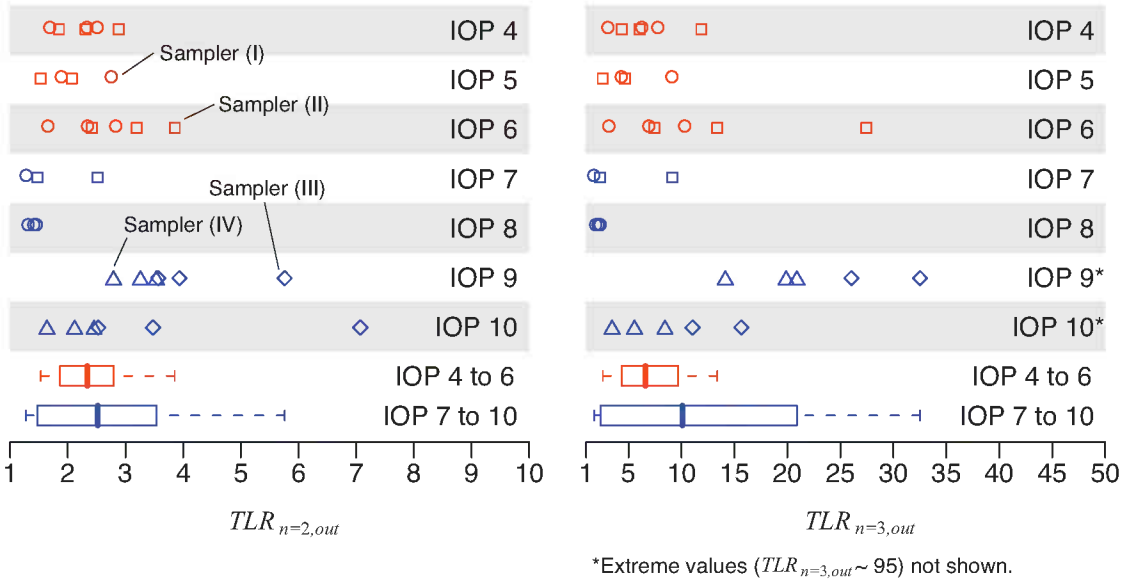

Figure 1. Toxic load ratio (*TLR*) computed from outdoor concentrations for $n = 2$ (left) and 3 (right). There are up to three releases in each IOP where the outdoor concentrations were measured at two locations. The boxplots summarize the lower quartile, median, and upper quartile *TLR* for the daytime (IOPs 4–6) and nighttime (IOPs 7–10) releases. Boxplot whiskers show extent of *TLR* excluding the outliners.

The values of TLR_{out} shown in Figure 1 are more variable during the nighttime releases. This is, in part, because of larger fluctuations in concentrations during daytime releases than during nighttime [16]. Additionally, the release location was much closer to the study building during the evening experiments, 50 m away for IOPs 9 and 10, as opposed to 100 to 300 m for IOPs 7 and 8 (see Table 1). Finn et al. [16] reported a general trend that the tracer gas concentrations were more variable at shorter distances from the release source.

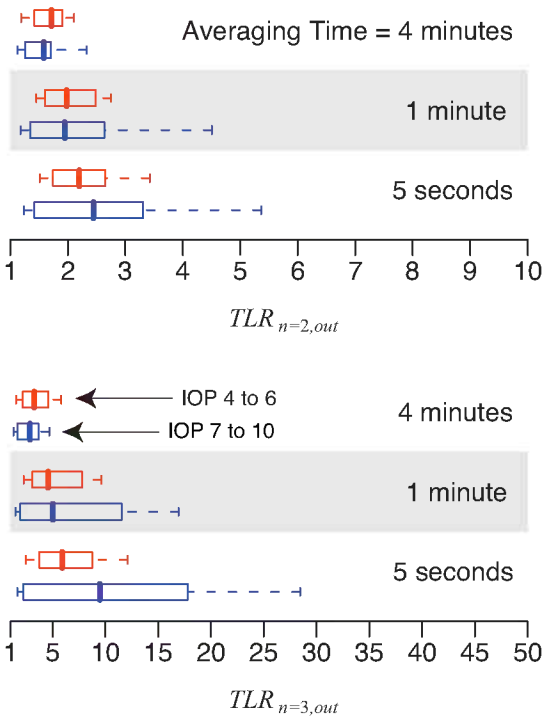

Figure 2. Boxplots of *TLR* where the toxic load is computed from different time aggregations of outdoor concentrations. Results are shown for $n = 2$ (top) and 3 (bottom), and with daytime releases (IOPs 4–6) shown separately from the nighttime releases (IOPs 7–10).

Figure 2 shows TLR_{out} for $n = 2$ and $n = 3$ calculated from outdoor concentrations at an averaging time of 5 seconds, 1 minute, and 4 minutes. The unedited tracer gas data have a time resolution of 0.5 seconds for the TGAs (samplers I and II), and one second for the Mirans (samplers III and IV). As expected, longer averaging times produce lower toxic load estimates. However, compared to the boxplots shown in Figure 1, the change in TLR_{out} is negligible when a 5-second time averaging interval is used. This implies concentration fluctuations at a timescale finer than 5 seconds will have negligible impact on the estimates of toxic load. This is important because 5 seconds is a little longer than the typical duration of a human breath. Time averaging of minutes might, however, lead to an underestimation of TLR_{out} by a factor of 2 to 5, assuming that the functional form used to calculate toxic load, i.e., Eq. (1), is valid.

4.2. Effects of Time Aggregation on SIP Effectiveness

To determine the effects of time aggregation on SIP effectiveness, we first computed *IO* as defined in Eq. (3a) as the base SIP scenario where $\overline{C}$ is used to compute *TL*. The outdoor toxic load was calculated using the average concentrations measured at the two rooftop air intakes. The indoor toxic load was calculated for the lower floors that are served by the fifth floor mechanical room, and then separately for the upper floors served by the rooftop mechanical room. The time period considered was approximately one hour from the start of the release, as described in Table 2.

Figure 3 shows the resulting *IO* for different values of *n*. When $n = 1$ (i.e. adverse health effects are predicted by the cumulative exposure), the median *IO* was 0.73 for the upper floors and 0.26 for the lower floors. *IO* was lower in the lower floors because the transport of tracer gas into the building was predominantly through the rooftop air intakes. As the value of *n* increased, *IO* decreased because the exponent magnifies the difference between indoor concentrations that were lower relative to outdoor concentrations. When $n = 2$, the median *IO* for the upper floors was 0.54. When $n = 3$, the median *IO* was 0.40. For the lower floors, the median *IO* was 0.05 and 0.01 when $n = 2$ and 3, respectively.

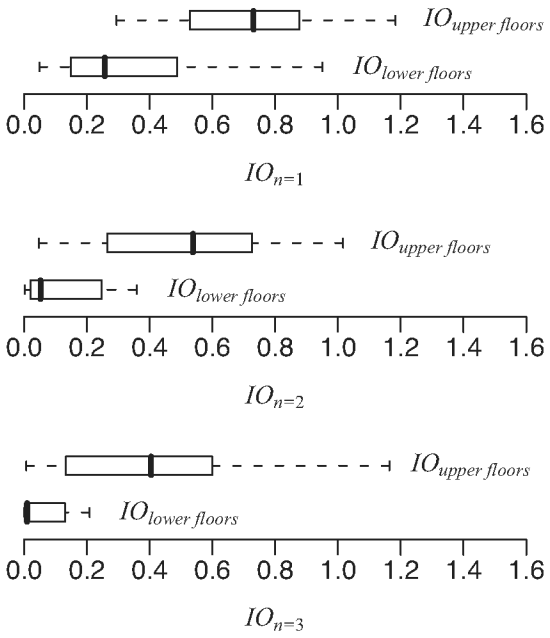

Figure 3. Ratios of the indoor to outdoor toxic load (IO) computed from the time-averaged concentrations measured at Building A. Outdoor concentrations were measured at the rooftop air intakes. Indoor concentrations were measured in the upper and lower floors. Results from all IOPs 4–10 are shown for n = 1 (top), 2 (middle), and 3 (bottom).

In all cases, SIP was always effective and preferable to staying outdoors. There are only two instances where IO was greater than one: the second release during IOP 7 and the third release of IOP 10. In both cases, tracer gas measured at the rooftop air intakes had substantially lower concentrations compared to the previous release(s) during the IOP. This implies that some of the tracer gas found indoors was likely residue from previous release(s). These two cases were excluded from the further analysis that follows.

When n does not equal one, IO will differ depending on the levels of time aggregation used to compute the outdoor and indoor toxic load. As defined in Eq. (3b), we computed Φ using the 4-minute concentration data by first calculating the values of TLR_{out} and TLR_{in}. Because the outdoor concentrations are generally more variable than the indoor concentrations, the value of Φ is less than one. Relative to the base case where IO was computed from time-averaged

concentrations, using the 4-minute data instead would reduce *IO* by a factor of Φ. Figure 4 shows that the median value of Φ is approximately 0.5 when $n = 2$, and 0.25 when $n = 3$. This would imply that by using the 4-minute concentration data, SIP effectiveness would be twice as effective when $n = 2$, and four times as effective when $n = 3$, relative to the base case computed from time-averaged concentrations.

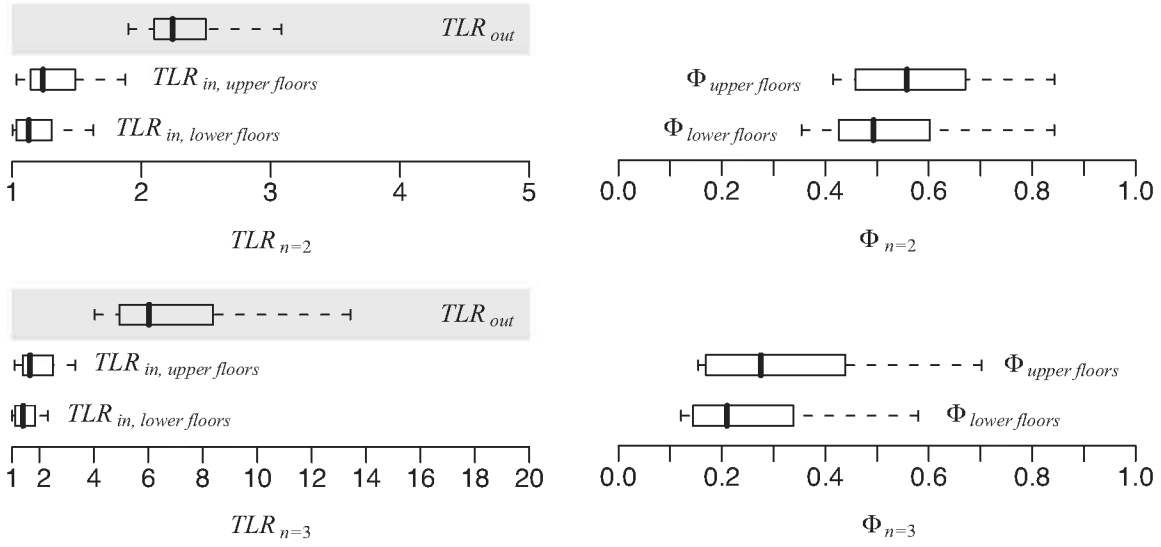

Figure 4. Toxic load ratio (*TLR*) and Φ calculated from the ratio of TLR_{in} and TLR_{out} (see Eq. (3b)). The outdoor and indoor toxic loads are computed using 4-minute concentration data measured at Building A. Results from all IOPs 4–10 are shown for $n = 2$ (top) and 3 (bottom).

If tracer gas concentrations were monitored at time intervals less frequent than 4 minutes, we would expect outdoor concentrations measured at the rooftop air intakes to vary more rapidly with time than the concentrations measured inside the mechanical rooms of the building. We do not have actual data to demonstrate this, but it is possible to compute a rough estimate on how this might impact Φ by referring to Figure 2. TLR_{out} computed from the tracer gas concentrations measured by the outdoor fast-response analyzers is about a factor of 1.4 higher when the averaging time is 5 seconds, relative to 4 minutes, when $n = 2$. This would imply that had the

concentrations measured at the rooftop air intakes been available at a 5-second time resolution, the Φ would further decrease by a factor of approximately 1.4 from the results shown in Figure 4. Similarly, for $n = 3$, Φ would decrease further by a factor of two. In other words, the computed Φ would mean that SIP effectiveness is about three times as effective when $n = 2$, and eight times as effective when $n = 3$, than the case where toxic loads are calculated from time-averaged concentrations.

5. DISCUSSION

5.1. Uncertainty Introduced by Time Averaging Intervals

For $n = 2$, toxic load may differ by a factor of six at the most, depending on the levels of time aggregation used in computation. This limited range of influence from time aggregation on toxic load was observed in a wide variety of experimental conditions, including daytime and nighttime releases, HVAC system on and off, and indoor concentrations measured in different parts of the building. At higher n, the difference caused by time aggregation is only somewhat larger. For example, if the appropriate timescale for computing the toxic load of a chemical is 5 seconds, then the use of 4-minute concentration data would lead to an underestimation of the toxic load by a factor of no more than 30 for $n = 3$ (Figure 2). Although this might appear to be a substantial difference, we have results only for experiments where the toxic load was computed from samplers located very close to the source. We would expect lesser differences when the building is further away from the release location. More importantly, Figure 4 shows that the indoor toxic loads are much less affected by the choice of time aggregation compare to the outdoor toxic loads.

For evaluating SIP effectiveness, the time resolution of the concentration data—and its resulting impact on the toxic load calculation—has very limited effect on IO when $n = 2$. When $n = 3$, SIP could be four to eight times as effective, depending whether concentration data on the order of minutes or seconds were used to compute toxic loads.

5.2. Overall Uncertainty in Estimates of Toxic Load and SIP Effectiveness

To gain perspective on the impact of time aggregation intervals, it is useful to consider other sources of uncertainty in calculating toxic load and SIP effectiveness. Such comparison will allow us to judge if the method used to compute toxic load is the dominant source of uncertainty in a SIP assessment or if there are other sources of uncertainty that are greater contributors. Accordingly, we consider two other sources of uncertainty: thresholds for acute exposure and outdoor concentration predictions.

AEGLs are developed to assess the hazard of brief exposures to high concentrations. Currently, AEGLs are available for 48 high-priority chemicals of acute toxicity [15]. Uncertainty factors, or margins of safety, are commonly applied when extrapolating from animal data to estimate human risk levels. A ten-fold interspecies uncertainty factor is generally applied when extrapolating from results of studies on experimental animals to humans. When quantitative data on a sensitive subpopulation are lacking, another ten-fold uncertainty factor is applied to account for the variable sensitivity to humans, such as the differences between infants and adults. This intraspecies uncertainty factor is also applied when a broad spectrum of effects has been observed. Thus, a total uncertainty factor of 100 is often applied when deriving many AEGLs. In some cases when the interspecies and/or intraspecies toxicity differences appear to be small, the uncertainty factor is reduced from 10 to 3. The uncertainty factor applied to AEGLs is therefore a minimum of 10 (3×3). More commonly, however, the minimum uncertainty factor is at least

30. Consequently, the uncertainty introduced by the levels of time aggregation used to compute toxic load (six-fold at most) is smaller than the uncertainty factor in deriving AEGLs.

Uncertainty associated with the predictions of outdoor concentrations can be substantial as well. This is particularly true in urban areas due to complex flow around buildings [1]. Besides JU2003, there have been three other major U.S. urban field tracer experiments in the past decade. Hanna et al. [1] performed simulations of these experiments using the Hazard Prediction Assessment Capability (HPAC)–Urban and the Joint Effects Model (JEM) transport and dispersion models. They compared 30-minute average concentrations predicted by the models to the concentrations measured at various ground-level outdoor air samplers. Overall, about one- to two-fifths of the model-to-data comparisons differed by a factor of two or less. The comparisons are likely to be in worse agreement at shorter time averages. The choice of the time averaging interval for the toxic load estimates would add to the overall uncertainty of the outdoor assessment. But if the ultimate goal is to evaluate SIP effectiveness by way of an indoor-outdoor comparison, our analysis shows that the levels of time aggregation are generally not a major source of uncertainty in the overall assessment.

6. CONCLUSIONS

It remains a subject of debate how toxic load should be calculated for time-varying concentrations. In the absence of toxicological data, there is a need for simple methods to compute toxic loads for hazard assessment and to inform emergency response. Many have argued that the way toxic load is defined in Eq. (1) is unfounded and have proposed alternatives [5-7], including consideration of additional timescales such as the uptake time constant of a chemical for a given response [7], or the functional form of the recovery process [6]. However,

formulating such toxic load models would create an even greater demand for chemical-specific toxicological data. The sheer number of chemical- and health-end-point specific combinations required for toxicological testing is prohibitive for this question to be addressed experimentally. While pharmacokinetic modeling might be a viable alternative [17], it requires a detailed understanding of the mechanistic effects of chemical toxicology to the target organs, where information is also limited.

In this analysis, we considered a simple way of computing toxic load that depends only on concentration data $C(t)$ and the exponent n. The analysis of the JU2003 experiments show relatively minor differences in the computed toxic load due to time averaging compared to other sources of uncertainty in hazard assessment. For example, as reflected in how the AEGLs are established, toxicological data are highly uncertain. In addition, there are many unknowns within a typical hazard assessment, such as details of the release, local weather conditions, transport of the chemical into building and its fate, building operating conditions, and human susceptibility to the chemical among the general population. In comparison to these sources of uncertainty, the time averaging used to compute TL introduces only a small source of error to SIP effectiveness assessment.

For this reason, it is reasonable for health assessors to apply time-resolved modeling predictions to evaluate health effects, even when the exposure-response relationship for fluctuating concentrations remains unknown. For the purpose of computing toxic load and evaluating SIP effectiveness, a practical approach is to choose an averaging time interval that is comparable with the reference exposure limits, such as AEGLs. Hazard assessment models can utilize this simple rule-of-thumb to evaluate the toxic effects of hazardous chemical releases.

Further analysis of other urban-scale field experiments is needed to consider a wider range of release scenarios and building types. This analysis is based on data from one set of urban field experiments. The results presented here may not apply to cases where the outdoor ambient conditions are very different from the JU2003 experiments. The building considered here is a reasonable representation of a typical mid- to large-size office building that is equipped with a mechanical system and rooftop air intakes. But the results discussed here may not be representative of other building types. Further evaluation is required to see if the findings apply more broadly to other release scenarios involving different types of buildings.

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REFERENCES

[1] S. Hanna, J. Chang, I. Sykes, Evaluation of HPAC-Urban and JEM with U2000, JU2003, MSG05, and MID05 Tracer Observations, in: 91st American Meteorological Society Annual Meeting, Seattle, WA, 2011.

[2] W.R. Chan, W.W. Nazaroff, P.N. Price, A.J. Gadgil, Effectiveness of urban shelter-in-place--I: Idealized conditions, Atmos. Environ. 41 (2007) 4962-4976.

[3] W.R. Chan, W.W. Nazaroff, P.N. Price, A.J. Gadgil, Effectiveness of urban shelter-in-place--II: Residential districts, Atmos. Environ. 41 (2007) 7082-7095.

[4] W. Chan, W. Nazaroff, P. Price, A. Gadgil, Effectiveness of urban shelter-in-place--III: Commercial districts, Build. Simul. 1 (2008) 144-157.

[5] S.A. Stage, Determination of acute exposure guideline levels in a dispersion model, J. Air Waste Manage. Assoc. 54 (2004) 49-59.

[6] H. Kaplan, A model for the toxic dose under time-varying concentration, J Hazard. Mater. 167 (2009) 351-356.

[7] T.L. Hilderman, S.E. Hrudey, D.J. Wilson, A model for effective toxic load from fluctuating gas concentrations, J. Hazard. Mater. 64 (1999) 115-134.

[8] D.J. Ride, A practical method of estimating toxic loads in the presence of concentration fluctuations, Environmetrics 6 (1995) 643-650.

[9] USEPA, Acute Exposure Guideline Levels (AGELs) Program, in: http://www.epa.gov/oppt/aegl/, accessed 2012.

[10] W.F. ten Berge, A. Zwart, L.M. Appelman, Concentration–time mortality response relationship of irritant and systemically acting vapours and gases, J. Hazard. Mater. 13 (1986) 301-309.

[11] K.J. Allwine, J.E. Flaherty, Joint Urban 2003: Study Overview and Instrument Locations, in: Pacific Northwest National Laboratory, Richland, Washington, 2006, pp. 92.

[12] D. Black, R. Sextro, T. Thatcher, W. Delp, S.C. Chang, E. Wood, J. Deputy, T. Hotchi, M. Sippola, D. Sullivan, Joint Urban 2003: Indoor Measurements Final Data Report, in: LBNL Report, Lawrence Berkeley National Laboratory, Berkeley, 2004, pp. 13.

[13] S.T. Parker, C.J. Coffey, Analytical solutions for exposures and toxic loads in well-mixed shelters in support of shelter-in-place assessments, J. Hazard. Mater. 192 (2011) 419-422.

[14] S. Hanna, J. White, Y. Zhou, Observed winds, turbulence, and dispersion in built-up downtown areas of Oklahoma City and Manhattan, Boundary Layer Meteorol. 125 (2007) 441-468.

[15] NRC, Acute Exposure Guideline Levels for Selected Airborne Chemicals, The National Academies Press, Washington, DC, 2007.

[16] D. Finn, K.L. Clawson, R.G. Carter, J.D. Rich, C. Biltoft, M. Leach, Analysis of Urban Atmosphere Plume Concentration Fluctuations, Boundary Layer Meteorol. 136 (2010) 431-456.

[17] M.V. Evans, W.K. Boyes, J.E. Simmons, D.K. Litton, M.R. Easterling, A comparison of Haber's rule at different ages using a physiologically based pharmacokinetic (PBPK) model for chloroform in rats, Toxicology 176 (2002) 11-23.