Measuring urban bicyclists' uptake of traffic-related volatile organic compounds using ambient and breath concentrations

Alexander Y. Bigazzi
Email: abigazzi@pdx.edu

Wentai Luo
Email: wentai@cecs.pdx.edu

Miguel A. Figliozzi
Email: figliozzi@pdx.edu

James F. Pankow
Email: pankowj@pdx.edu

Lorne M. Isabelle
Email: lorne@cecs.pdx.edu

Portland State University
Department of Civil and Environmental Engineering
P.O. Box 751
Portland, Oregon 97207-0751, USA
Phone: 503-725-4282
Fax: 503-725-5950

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ABSTRACT

Urban bicyclists' uptake of traffic-related air pollution is still not well quantified, due to a lack of direct measurements of uptake and a lack of analysis of the variation in uptake. This paper describes and establishes the feasibility of a novel method for measuring bicyclists' uptake of volatile organic compounds (VOC) by sampling breath concentrations. An initial data set demonstrates the ability of the proposed method to generate findings for transportation analysis, with statistically significant exposure and uptake differences from bicycling on arterial versus bikeway facilities for several traffic-related VOC. Breath concentrations of toluene, ethylbenzene, m-xylene, and o-xylene increased at 5-10% of the increase in exposure concentrations, which were more than 2 times higher on arterials than bikeways. The elasticity of breath to ambient concentration increases for these VOC ranged from 0.19 to 0.54, and breath concentrations were 19-45% higher after bicycling on arterials than after bicycling on bikeways. These results provide the first empirical evidence that the usage of bikeways (or greenways) by bicyclists within an urban environment can significantly reduce uptake of dangerous traffic-related gas pollutants. Dynamic concentration and respiration data reveal unfavorable correlations from a health impacts perspective, where bicyclists' respiration and travel time are greater at higher-concentration locations on already high-concentration roadways (arterials).

1 INTRODUCTION

Urban transportation systems can affect traveler health in many ways. Bicyclists and other active travelers enjoy the health benefits of increased physical activity *(1)*, but with the potential drawbacks of increased crashes and uptake of traffic-related air pollutants *(2, 3)*. It is clear from past research that exposure to traffic-related air pollution has negative health impacts for urban populations *(4)*, and exposure during travel can be especially dangerous because of proximity to sources of pollution *(5–7)*. However, the details of exposure concentrations within individual transportation microenvironments are not well established because of the great diversity of environmental, meteorological, and traffic conditions *(8, 9)*. The health risks of pollution exposure during bicycling are particularly uncertain because of varying levels of physical exertion that affect the uptake of pollutants through varying volumes and depths of respiration *(10, 11)*. The current state of uncertainty about bicyclists' uptake of traffic-related air pollution leaves unsatisfying gaps in health impact assessments and impedes health-conscious transportation planning and management.

1.1 Framework

The pathway of traffic-related pollution exposure, from motor vehicle emissions to health effects, is illustrated in Figure 1 – adapted from *(12)*. Motor vehicle emissions (Figure 1a) degrade urban air quality (Figure 1b) in accordance with atmospheric dispersive, chemical, and physical processes *(13)*. Travelers' exposure concentrations (Figure 1c) then depend on the space-time path of their travel *(14)*. The inhalation of traffic-related air pollution (Figure 1d) depends on travelers' breathing volume while exposed to a pollutant concentration. Uptake of the inhaled pollutants into the body (Figure 1e) depends on physiological processes in the human respiratory and circulatory tracts *(15–17)*. Finally, the health effects (Figure 1f) of air pollution uptake doses is a function of the toxicity of the pollutants and physiology of the individual *(4, 16, 18, 19)*. The present research only addresses the steps from exposure concentrations to uptake doses (Figure 1c to 1e). This research does not explicitly model emissions or the atmospheric dispersion and chemical transformations that lead to ambient concentrations. Similarly, health outcomes of uptake doses are left for future study.

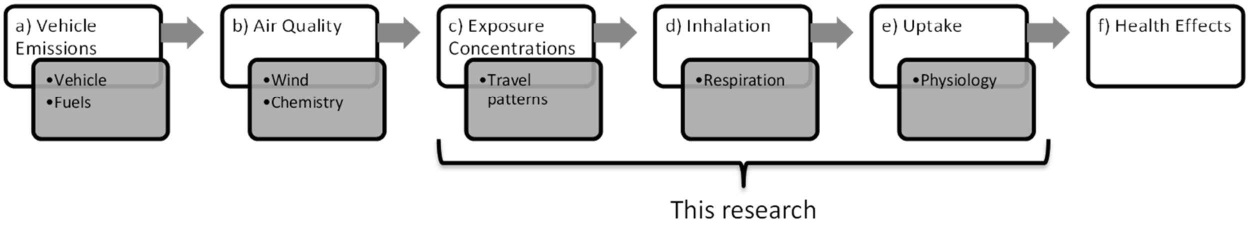

Figure 1. Exposure Pathway of Traffic-Related Air Pollution from Emissions to Health Effects

1.2 Literature Review

Some research has shown that bicycle facilities can affect bicyclists' pollution exposure concentrations, generally by creating distance between bicyclists and motor vehicles *(20–22)*. The more robust traveler exposure studies apply different respiration rates for travelers of different modes *(23–25)* or even on different trips *(26)*, but intra-modal variability in respiration is generally ignored. In the transportation literature respiration is almost never a function of travel or roadway characteristics other than mode. One exception is McNabola et al. *(27)*, who used speed-varying respiration rates for a bicyclist, though the respiration model is static and not related to the transportation network. A second exception is Int Panis et al. *(28)*, who measured on-road respiration and used transient respiration rates to estimate dynamic pollution intake.

Only two studies have directly measured bicyclists' uptake of traffic-related air pollution. Bergamaschi et al. *(29)* found significant increases of benzene and toluene in blood for bicyclists in urban areas and significant increases of toluene and xylenes in urine. Although uptake was directly measured, respiration was not. No significant changes in blood or urine concentrations of VOC were found for rural bicyclists, and no further variation in urban bicycling was tested. Nwokoro et al. *(7)* measured inhaled doses of black carbon (BC) by bicyclists in London by sampling airway macrophages in "induced sputum". They found significantly (63%) higher inhaled doses of BC for bicyclists than non-bicyclists, correlated with higher commute exposure concentrations. Bicyclists also had almost twice as long commute durations, and experienced 41% of daily BC exposure during the commute (as compared to 19% for non-bicyclists).

Two additional studies modeled uptake of volatile organic compounds (VOC) using a Human Respiratory Tract Model (HRTM) from the International Commission on Radiological Protection (ICRP) *(16)*. McNabola et al. *(27, 30)* used both on-road measured and assumed exposure concentrations with laboratory-measured respiration characteristics to estimate lung deposition of fine particulate matter ($PM_{2.5}$) and absorption of VOC for different travel modes using the HRTM. They found that uptake does not follow the same modal patterns as differences in exposure concentrations. Bicyclists had the highest total lung deposition of $PM_{2.5}$ and the second-highest absorption of VOC over similar trips to other modes. Travelers' breathing affected the intake dose and the location of absorption, with more benzene absorbed in the alveolar-interstitial region (deep in the lungs) for bicyclists and pedestrians, and more benzene absorbed in the extrathoracic region (near the mouth and nasal passages) for car passengers. Breathing pattern affected benzene absorption more than 1,3-butadiene absorption because of benzene's lower solubility (almost all 1,3-butadiene was absorbed in the extrathoracic region).

These few existing studies of bicyclists' uptake suggest that exposure concentrations can be poor surrogates for uptake doses, though the differences between intake and uptake doses have yet to be analyzed. Also, varying uptake on different parts of the transportation network has not been analyzed.

1.3 Objectives

To date, VOC uptake by bicyclists has only been directly measured in one known study, and only urban versus rural riding was tested. Additionally, there have been few concurrent on-road measurements of bicyclists' exposure concentrations and respiration – and no such research has incorporated detailed travel data. McNabola et al. model VOC uptake differences by travel mode *(27)* and by bicycling speed *(30)*, but both studies use laboratory-based physiological parameters for the model and the assumed breathing is independent of the travel environment (neglecting the effects of grade, for example). Thus, two main gaps in the literature related to bicyclists' uptake of traffic-related VOC are a lack of direct measurements of uptake and a lack of analysis of the variation in uptake within an urban environment.

The objectives of this paper are to:

1. describe a novel method for directly measuring bicyclists' uptake of traffic-related VOC using breath samples,
2. establish the feasibility of the proposed method for measuring uptake, and
3. demonstrate the ability of the proposed method to generate findings for transportation analysis, with results from the initial data set.

The next section describes the uptake and exposure measurement methods.

2 METHODOLOGY

This section describes 1) the proposed uptake measurement method 2) instrumentation, and 3) data collection procedures.

2.1 The proposed uptake measurement method

The proposed method for measuring bicyclists' uptake of VOC is to collect end-tidal breath samples before and after bicycling on segments of a route. Breath analysis of VOC is increasingly used as a non-invasive method for measuring health biomarkers *(31)*. The exhaled air is representative of concentrations in the alveolar air, which can reasonably be assumed to be in equilibrium with the blood (with blood and alveolar air concentrations at the ratio of the blood/gas partition coefficient) *(32)*. Thus, breath concentrations represent VOC absorption into the blood, which is the first and most dynamic body "compartment" for inhaled VOC *(17)*. Although these techniques are established in the medical and exposure science fields, they have not been applied to studies of travelers.

Bicyclist breath samples are collected in the field using gas sample bags with a mouthpiece attached. End-tidal breath is needed because dead space air contained in whole-breath samples will less precisely represent the alveolar air. Excluding the first 200 mL of an exhalation will generally avoid dead space air, and exhalation sampling is generally not affected by the exhalation pattern used *(31)*. The sampling procedure in this study is to capture only the second half of an exhaled breath in the sample bag (in order to avoid dead space air), allowing multiple breaths per sample bag as needed.

After riding, the bags are immediately brought back to a laboratory and the breath samples are extracted by drawing the air through stainless steel adsorption/thermal desorption (ATD) cartridges with a dual sorbent bed (Tenax TA and Carbophaph 1TD). The samples are then thermally desorbed and analyzed using gas chromatography followed by mass spectrometry (GC/MS) to assess concentrations of VOC, as described in *(33–35)*. The instruments used for gas analysis are a PerkinElmer TurboMatrix 650 adsorption/thermal desorption (ATD) unit as interfaced to an Agilent 5975C GC/MS. Standards are prepared and used to calibrate the instrument response. Ambient VOC concentrations during riding are also sampled and analyzed using ATD cartridges and GC/MS, as described in the next section.

A series of pilot data collections were undertaken to establish the feasibility of the proposed method for measuring bicyclists' uptake of VOC while riding. Minor modifications to the method were made based on the early results, as described below in the Results section. An initial data collection period followed, using the instrumentation described in the next section. A more extended data collection period based on this methodology is ongoing.

2.2 Instrumentation

On-road data collection requires several devices to be simultaneously mounted on a test bicycle and assiduously synchronized. The on-road data streams are:

1. Travel data monitoring

   The position of the bicyclist is measured with three redundant Global Positioning System (GPS) receivers recording 1 Hz location data with time stamps. Redundant GPS sensors are used to cross-check the location data for reliability. Additional measurements are made of the bicycle speed and acceleration.

2. Physiology monitoring (respiration and heart rate)

   Respiration rate is measured using the Zephyr BioHarness 3 (Figure 2), which can collect and log 1-Hz physiology data for several hours. The BioHarness is worn by the bicyclist around the chest, next to the skin. It uses a conductive elastic polymer strap that can measure expansion of the chest to detect respiration frequency. A breathing amplitude is also measured, which is proportional to the expansion of the chest band. Thus, tidal volume is not directly measured, but can be inferred from breath amplitude.

3. Ambient/exposure VOC monitoring

   Two different methods are used to measure VOC exposure concentrations. A near-continuous (1 Hz) device measures concentrations of total volatile organic compounds (TVOC) at the same scale as the high-resolution travel data obtained through GPS. At the same time, ATD cartridges are used to collect time-averaged VOC samples over ride segments of 20-30 minutes (which are then analyzed for concentrations of many individual VOC). The individual compound analysis is a more precise and detailed method of analysis than measuring TVOC, but can only provide low-resolution data (10 minutes or more), and so is insufficient, by itself, to address research questions about highly space-resolved air quality. Combining the two methods (time-averaged gas sampling and near-continuous TVOC measurements) provides detailed information about both the composition and time-space variation of VOC concentrations, and is one of the novel aspects of this research.

   Near-continuous TVOC concentrations are measured using the IonScience PhoCheck Tiger. The Tiger measures TVOC using a photoionization detector (PID) with a 10.6 eV lamp, which detects compounds with an ionization potential below 10.6 eV. Individual compounds within that range are not distinguished, and the reported concentrations are in isobutylene-equivalent units. The Tiger measures a TVOC concentration range of 1 ppb to 20,000 ppm, with a resolution of 1 ppb. The Tiger is lightweight (0.72 kg) and portable, capable of operating on battery power for over 4 hours while collecting 1 Hz measurements. The data collections occurred within the annual factory calibration period and before 100 hours of use (when re-calibration is needed). The instrument is zeroed with a carbon filter at the beginning and end of each collection (which takes place in a clean environment), and a convex piecewise linear zero reference curve is applied. The Tiger is a new model of portable PID within the IonScience PhoCheck line, and so has not yet been used in published studies, to our knowledge. Earlier models of the PhoCheck were used for air quality studies in motor-vehicle environments *(36–38)*.

Time-averaged samples of ambient (exposure) volatile organic compounds (VOC) are collected simultaneously with the Tiger-TVOC measurements using stainless steel ATD cartridges with a dual sorbent bed and SKC personal sampler pumps. The samples are then desorbed and analyzed using the same thermal desorption and GC/MS method described above (Section 2.1). The sorbent cartridge with GC/MS method is common practice for outdoor air quality sampling *(33)*, though some roadside and on-road studies have used whole-air canister samplers *(39, 40)* or Tedlar bags *(27)* instead of sorbent cartridges to collect the sample.

4 Breath concentrations of VOC

Bicyclists' end-tidal breath samples are collected before and after riding on each test segment using 3-L FlexFilm™ bags from SKC (Figure 2). Immediately after data collection the breath samples are returned to the lab and extracted from the bags and analyzed using thermal desorption and GC/MS as described above (Section 2.1).

Figure 2. Breath sample bag (left) and physiology monitor strap (right)

2.3 Data Collection

A pilot study period ran from November 2012 through February 2013 in which different breath sample bags, start locations, and data collection procedures were tested to develop a sound and consistent data collection methodology. A validation data collection occurred April 2nd, 2013 with repeated measurements to establish the feasibility of measuring changes in breath concentrations. Three data collection days in July 2013 (2nd, 9th, and 11th) applied the developed method to measure uptake differences on different roadway facilities. Results are presented below for the April and July data collection days. A longer data collection period applying the described methodology is ongoing. In the pilot and initial data collections a single subject is used throughout to avoid the impact of differences in individual physiology on uptake.

On-road data were collected in Portland, Oregon on a mix of roadway facilities. The data collection route for the July 2013 data collection days is shown in Figure 3. The start location is in Mt. Tabor park, which was determined in the pilot phase to have sufficiently low ambient VOC concentrations. The first ride segment follows bikeways (also known as neighborhood greenways) and local streets over a 6.8 km (4.2 mi) path; the next two ride segments travel along arterial streets, primarily SE Powell Boulevard, for 7.9 km (4.9 mi) in each direction. Powell Blvd. is a major arterial with annual average daily traffic of 21,000 to 35,000 vehicles along the test route, a high-frequency bus lines and regular peak-period congestion. Average daily traffic on the bikeways range from under 1,000 to 3,000 vehicles. All data collection days were clear and mild. The April data collection day took a similar route along Powell Blvd. but neglected most of the bikeway segment and continued into downtown Portland (to the west).

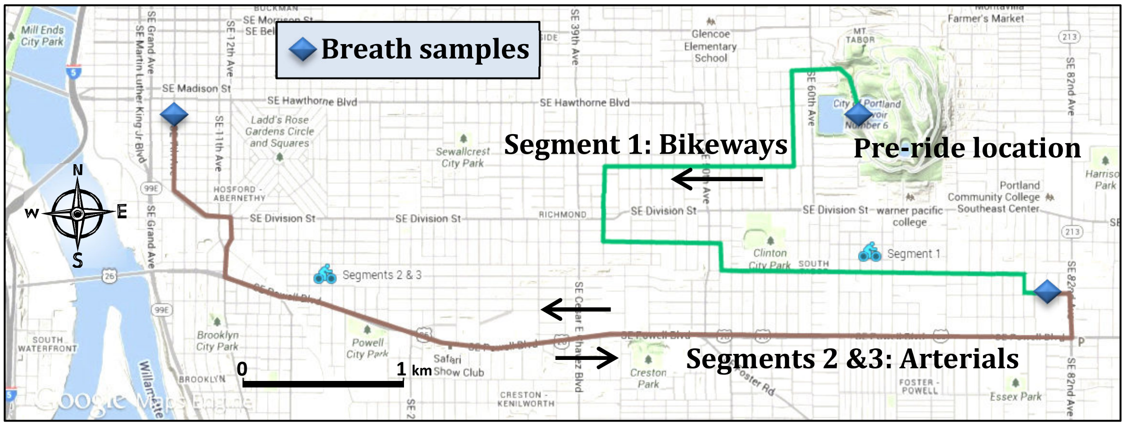

Figure 3. July 2013 data collection routes **(map imagery courtesy Google Maps)**

A pre-ride period of 30 minutes at the start location allows the bicyclist to equilibrate body concentrations with low ambient VOC concentrations. Time-average ambient samples are taken at the pre-ride location and along each ride segment using the SKC personal sampler pump (at 50 mL/min) and ATD cartridges. Breath samples are taken at the end of the pre-ride period and at the end of each ride segment. Each ride segment takes approximately 20-30 minutes to complete (providing 1-1.5 L ambient samples). The rider was instructed to ride cautiously and obey all traffic laws while riding at a moderate, comfortable pace. Data collections took place during morning peak periods, beginning around 7 am. The data collection bicycle with all equipment weighed 23.7 kg (52.2 lbs.).

3 RESULTS

3.1 Pilot Study Findings: Controlling Initial Conditions

A pilot study was undertaken to address potential problems in data and sample collection. The high variability of low-level VOC sampling presents several challenges for the proposed measurement approach. Low initial breath and blood concentrations of target compounds are needed to allow changes with uptake. Sampling residential, park, and on-road locations revealed that concentrations at urban residential off-road starting locations were too high for our study. Thus, experimental procedures include the subject remaining at a clean initial location (a park) for at least 30 minutes to equilibrate their blood concentrations with low background levels. This time period was considered sufficient because blood residence times are on the order of minutes for VOC *(17)*.

In addition, very low blanks are needed from the sample bags to achieve a sufficient signal/noise ratio. Several different bags were tested before finding a combination of bag material and valve that produced satisfactorily low blanks. Any valve lubricant must have very low VOC content - particularly for hydrocarbons, as the compounds of interest. Special arrangements were made with the bag manufacturer to obtain the needed materials. The bags can be cleaned and reused about 10 times before developing leaks that result in costly loss of samples.

High water content in breath is another challenge in analyzing breath samples. Water-soluble compounds can partition into condensed water in the bag if the sample temperate drops. Additionally, compounds that co-elute with water from GC column (such as acetone) have higher measurement variation. These considerations limit the compounds that can be reliably measured in breath. Despite these challenges, statistically significant differences were found when comparing ambient and breath samples taken after riding in distinct traffic conditions, as described below.

3.2 Feasibility of the uptake measurement Method

Samples collected from April 2013 show that a statistically significant increase of breath concentrations of certain traffic-related VOC can be measured with the proposed method. Five BTEX compounds (benzene, toluene, ethylbenzene, o-xylene, and m-xylene) were significantly higher in the post-ride breath at $p<0.10$, and all but benzene were significant at $p<0.05$. The exposure concentrations for these compounds during riding were 4.6 (for benzene) to 8.2 (for o-xylene) times higher than the pre-ride exposure concentrations, with average breath concentration increases of 20% (for benzene) to 65% (for m-xylene). The estimated measurement error (expressed as % root-mean-square error) based on the duplicate samples from April 2013 was highest for benzene (up to 38% for the pre-ride breath sample), but under 10% for most other target compounds.

3.3 Ambient and breath VOC concentration changes

The gas analysis results presented in this paper focus on a subset of 13 of the measured compounds that were present in all four days' breath samples at concentrations of at least 0.1 ng/L. Analysis of data from the three days in July with both bikeway and arterial riding reveals that VOC uptake during bicycling is strongly affected by facility type. Average changes in ambient (exposure) and breath concentrations of target VOC after riding on bikeways for 20-30 minutes (as compared with pre-ride concentrations) are shown in Figure 4. Significance levels for two-tailed paired t-tests are shown using '*' characters. Although there were some significant differences in ambient exposure concentrations, the breath concentrations do not change significantly (at $p<0.01$).

The concentration changes after riding on arterials (segments 2 and 3) as compared to the bikeway (segment 1) concentrations from July 2013 are shown in Figure 5. The ambient concentrations are much higher on the arterials than the bikeways, with statistically significant changes for most target compounds (again based on two-tailed paired t-tests). Carbon tetrachloride and CFC-113 are not traffic-related pollutants and are included as a reference, so an increase is not expected. Average concentration differences for the other compounds are 103-173% higher on the arterials. Breath concentrations increase an average of 5-57% for the traffic-related target compounds. The increases in breath concentrations of four BTEX compounds other than benzene are significant at $p<0.10$ or lower – the insignificant effect for benzene is likely related to the high breath sample variability described above in Section 3.2.

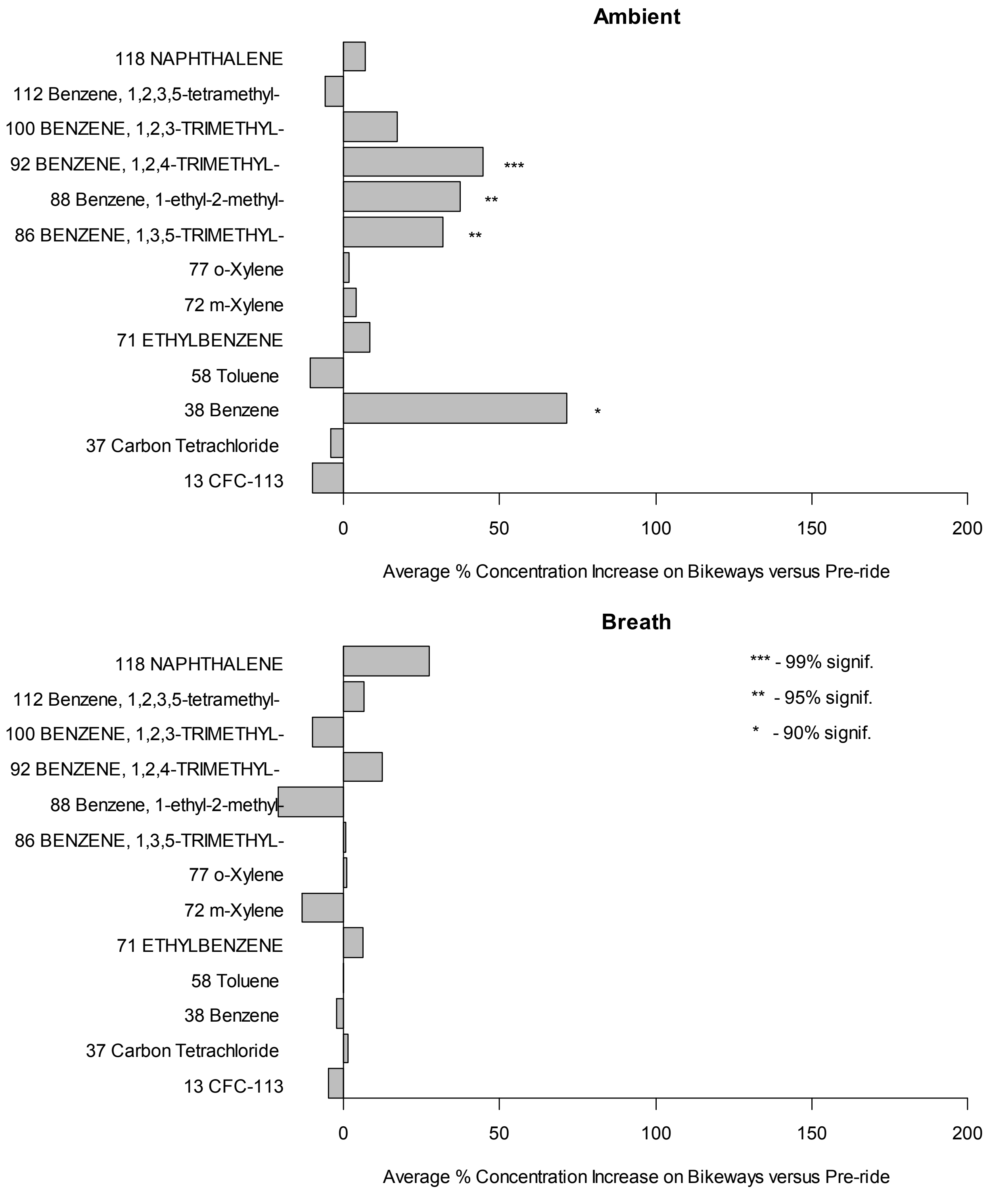

Figure 4. Average change in ambient and breath concentrations after riding on bikeways on July 2nd, 9th, and 11th (as compared with pre-ride concentrations)

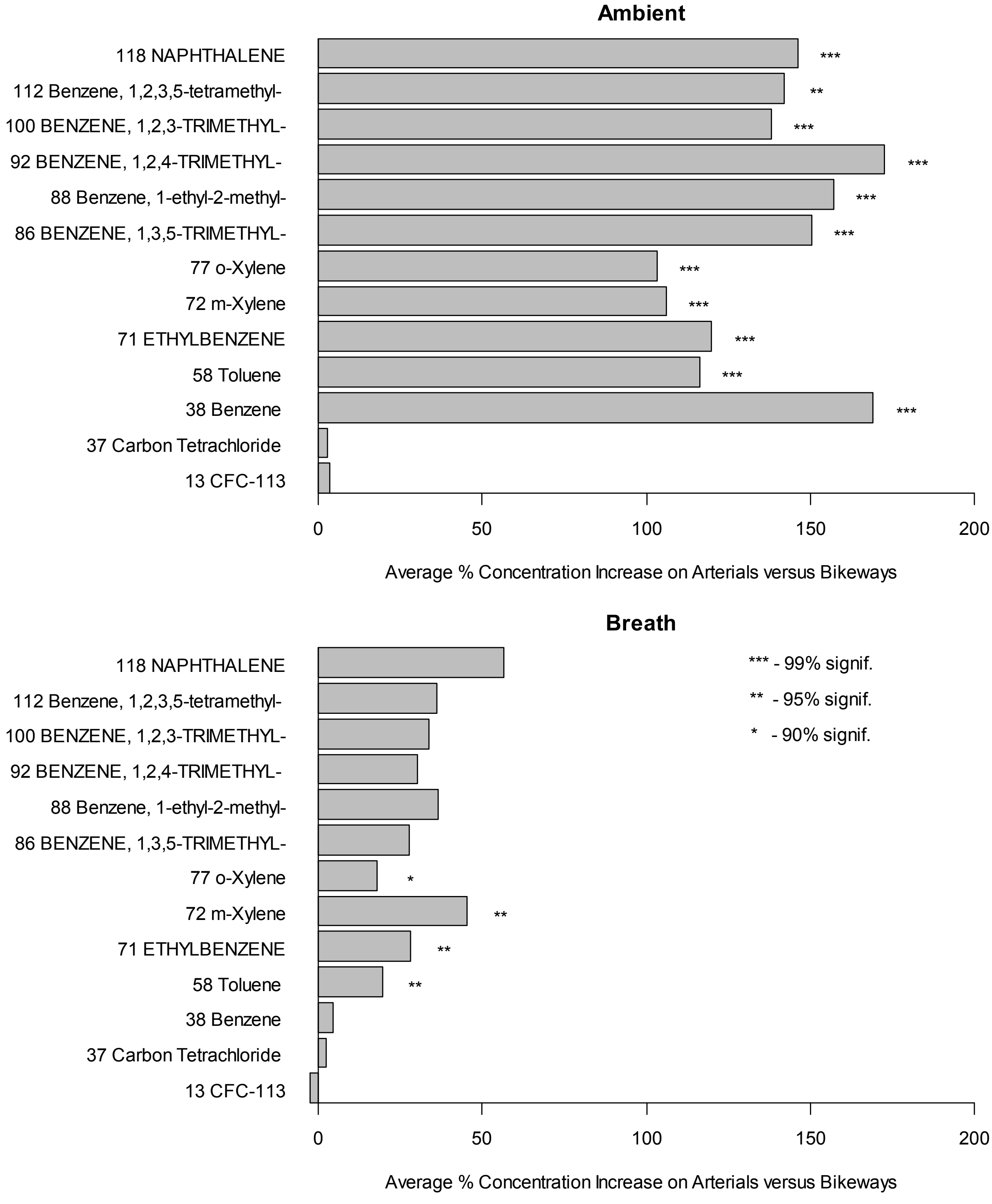

Figure 5. Average change in ambient and breath concentrations after riding on arterials on July 2nd, 9th, and 11th (as compared with bikeway concentrations)

The increases of ambient and breath concentrations for arterial versus bikeway riding were compared by regressing breath changes on ambient changes, excluding an intercept term. The ambient concentration change coefficient estimates for o-xylene, m-xylene, ethylbenzene, and toluene are all significant at $p<0.10$ with coefficients of 0.05 to 0.10. In other words, breath concentrations increased at around 5-10% of the ambient concentration increases for 20-30 minute rides. With breath concentrations after bicycling at 19-49% of ambient concentrations, the result of this increase is magnified 2-5 times in terms of percent change in breath concentrations – hence the 19-45% breath concentration increases for these compounds seen in Figure 5 with 100-120% ambient concentration increases. The average elasticity of breath to ambient concentration increases for these VOC was 0.19 to 0.54.

3.4 Dynamic exposure data

The measured uptake differences are caused by both exposure concentration and respiration changes. TVOC concentrations measured by the PID were on average 89% higher on the arterials than the bikeways (10.1 versus 5.4 in ppb-isobutylene) for data collected in July 2013. The second-by-second measurements also reveal more variability on the arterials, with standard deviations of 18.1 and 4.0 ppb-isobutylene on the arterials and bikeways, respectively. The 95th-percentile TVOC concentrations were 4.37 times the median value for arterials and 2.35 times the median value on bikeways.

Mean measured heart rate values were somewhat higher on the arterials than on the bikeways (92.2 versus 83.7 bpm), as were mean measured respiration rates (26.9 versus 26.2 breaths/min) and breath amplitudes (by a factor of 2.07). All three differences are statistically significant at $p<0.01$ based on two-tailed t-tests using the second-by-second data. The physiological differences probably relate to higher bicycle travel speeds on the arterials (19.3 versus 17.1 kph, or 12.0 versus 10.6 mph, statistically significant at $p<0.01$). Higher speeds could be due to operational characteristics (fewer stop controls on the arterial) or to subtler factors influencing the volitional speed (higher-speed passing motor vehicle traffic or stress or safety concerns). Another likely factor is a net loss of elevation on the bikeways (the arterial segments were closed-loop trips). Weather could also be a factor, as temperatures were significantly ($p<0.01$) higher during rides on arterials than bikeways, and higher on bikeways than at the pre-ride location (averaging 19.3, 16.7, and 15.1 °C on arterials, bikeways, and the pre-ride location, respectively).

Analysis of the second-by-second data also reveals small but significant ($p<0.01$) positive correlations between TVOC exposure concentrations and both heart rate and breath amplitude on the arterials ($r=0.03$ and $r=0.08$, respectively). These relationships are significantly *negatively* correlated on the bikeways. In addition, there is a positive correlation between travel speed and TVOC exposure concentration on the bikeways ($r=0.04$, significant at $p<0.01$) but a negative correlation between travel speed and TVOC exposure concentration on the arterials ($r=-0.02$, significant at $p=0.08$). A negative correlation between speed and TVOC concentration would mean that a bicyclist was delayed in higher-concentration areas – an unfavorable outcome for exposure risk. These early findings suggest that generally high-concentration roadways (arterials) also have operational characteristics that lead to unfavorable increases in travel time and respiration at locations where concentrations are higher.

4 CONCLUSIONS

This paper establishes the feasibility of a method for measuring uptake of VOC while bicycling in an urban setting. The method requires special low-blank gas sample bags and clean starting locations. Application of the method succeeded in finding statistically significant exposure and uptake differences after bicycling on arterial versus bikeway facilities for several traffic-related VOC. Breath concentrations of toluene, ethylbenzene, m-xylene, and o-xylene increased at 5-10% of the increase in exposure concentrations, which were more than 2 times higher on arterials than bikeways. The elasticity of breath to ambient concentration increases for these same VOC ranged from 0.19 to 0.54 – in other words a 10% increase in ambient VOC led to a 1.9% to 5.4% increase in breath concentrations. In terms of percent changes, breath (and by inference blood) concentrations of these traffic-related VOC were 19-45% higher after bicycling on arterials than after bicycling on bikeways and local streets – a significant increase in uptake. These results provide the first empirical evidence that the usage of bikeways (or greenways) by bicyclists can significantly reduce uptake of dangerous traffic-related gas pollutants compared with bicycling on arterial roadways.

Some of the uptake differences are potentially related to respiration differences by facility type. Dynamic concentration and respiration data reveal unfavorable correlations from a health impacts perspective, where bicyclists' respiration and travel time are greater at higher-concentration locations on already high-concentration roadways (arterials). These correlations could be due to a number of factors that simultaneously influence motor vehicle pollution emissions, motor vehicle proximity to bicyclists, bicyclist respiration rates, and/or bicycle travel speed (e.g. increase power requirements due to upward slopes or stop/acceleration activity at intersections, queuing during congestion).

The main objective of this research paper is to establish the feasibility of the methodology for measuring bicyclists' uptake. Next steps include measuring changes in breath concentrations for additional human subjects and comparing uptake between bicyclists riding in the same exposure conditions. Future research endeavors could assess the applicability of human uptake models for estimating bicyclists' uptake and determine rate constants for clearance of VOC after leaving high-concentration roadways. The integration of high-resolution traffic data may be useful to estimate the impacts of traffic characteristics on exposure. Analysis of detailed traffic and roadway data may also reveal some of the factors leading to the observed correlations between exposure concentrations and respiration.

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