

Response to Editor:

We thank the referees and editor for their helpful reviews. The manuscript has been revised accordingly with our responses interspersed below.

There are still a few remaining issues from the reviewers. Please address or clarify accordingly. Thanks.

(1) Figure 3b and associated text in lines 232-248: The ratio of the paint tracers to benzene is not systematically different on weekends compared to weekdays. What is the trend in concentrations? My assumption would be that benzene concentrations are similar on weekends to weekdays because automobile traffic typically has a small reduction on the weekends (compared to diesel, which falls significantly). But perhaps NYC is atypical in this regard and there are large reductions in vehicle traffic on the weekend.

We highlighted in the text that we might expect the coatings emissions to be smaller on the weekends compared to weekdays if industrial and/or construction activities reduced on the weekend, but this trend was not observed as shown in Figure 3b. It may even be true that these activities persist into the weekend, though data on coatings usage in NYC does not currently exist. As the reviewer suggested, the benzene concentrations only had a slight reduction from the weekdays to weekend days, with the lowest observed benzene concentration reported on Sunday. Because weekend benzene reductions were not significant, the ratios of the tracer compounds (PCBTF and Toluene) to benzene reflect changes in the usage patterns of coatings products that use these compounds as ingredients from weekdays to weekend days. The results suggest coating usage might not change by day of week in densely populated New York City, however, these usage trends might not reflect emissions in other U.S. cities. In order to draw more robust conclusions about weekend/weekday coatings usage, ambient measurements in other cities would be necessary. To better clarify these conclusions, the following text has been modified at L246:

“The mean concentration of benzene does not decrease significantly from weekdays to weekend days, therefore changes in the emission ratios to benzene by day of week should reflect changes in the usage of coatings products that use PCBTF and Texanol as ingredients. It might also be expected that coatings emissions are smaller on the weekends compared to weekdays, but this difference is not clearly observed in Fig. 3b and alternatively suggests that coating usage persists into the weekend in New York City, though these trends likely vary by city.”

(2) Figure 5 pie charts - for the SB polyurethane, it seems that aromatics are grouped into "HC". Please clarify.

The pie charts in Figure 5b show the distribution of VOC mass emitted and measured during each complete evaporation experiment. The largest 10 masses measured by the PTR-ToF-MS are labeled as the “most-likely” species. The PTR-ToF-MS measures the chemical formulae; however, isomers cannot be distinguished and explicit identification is not always possible. This

is especially true at low m/z , where large hydrocarbons ($>C_8$) fragment complicating spectra interpretation (Erickson et al 2014; Gueneron et al., 2015). For the solvent-borne polyurethane, the pie chart is labeled based on the formula at the detected mass (i.e. isoprene, cyclopentane fragments, and other fragments labeled as C_4 HC). The explicit identity of such a complex mixture of hydrocarbons that uses Stoddard solvent as its base is not possible using PTR-ToF-MS as the primary detection technique. Application of the GC as the front-end of the PTR even highlights the complexity of the mix of hydrocarbons in this product (alkanes, alkenes, cycloalkanes, aromatics, etc.). It is possible that aromatic fragments are included at some of the lower m/z , however, explicit identification is not possible. In order to clarify this, the following text is added to the Figure 5 caption:

“Several small hydrocarbon masses could not be explicitly identified due to fragmentation of larger masses and are instead labeled by their carbon number.”

Erickson, M. H., Gueneron, M., and Jobson, B. T.: Measuring long chain alkanes in diesel engine exhaust by thermal desorption PTR-MS, *Atmos. Meas. Tech.*, 7, 225–239, doi:10.5194/amt-7-225-2014, 2014.

Gueneron, M., Erickson, M. H., VanderSchelden, G. S., and Jobson, B. T.: PTR-MS fragmentation patterns of gasoline hydrocarbons, *Int. J. Mass Spectrom.*, 379, 97–109, doi:10.1016/j.ijms.2015.01.001, 2015.

(3) Figure 8a: A few sentences on how the CARB inventory calculates emissions would be warranted. Presumably the inventory emissions reflect the changing composition of paints and coatings over the period of 1990-2014.

CARB utilizes sales, formulation, and reactivity data from mandatory surveys conducted every few years. These data were used to calculate the quantity of total organic gases and total reactive gases (VOCs and LVP-VOCs). CARB then applies a fate and transport adjustment to ingredients to correct for other loss processes including down the drain or combustion as determined by a comprehensive review of the current literature. The data included in Figure 8a, contains survey data from five separate years (1990, 1998, 2001, 2005, and 2014) and reflects both the changing composition and usage (from sales) of coatings in California. L413-418 gave a brief description of the CARB inventory, however, we have re-worked the introductory paragraph of that section (L408) to improve the details of these emissions calculations:

“Every four to five years, CARB conducts comprehensive surveys of architectural coatings sold in California to gather information about the ingredients and sales with the goal of updating emission inventories. The response to the surveys is mandatory and CARB ensures the validity of the data following extensive quality assurance and quality control measures and the results accurately represent the sales volume in California and are made publicly available (CARB, 2018). These sales, product formulations, and reactivity data are combined together with fate and transport adjustments to account for non-atmospheric loss processes to estimate the total emissions of reactive organic gases.”

(4) It seems that one impact of this work is that it informs how paint and coating emissions can be represented in chemical transport models. Since most of the VOCs seem to be emitted very quickly (a few hours), does it mean that models can ignore longer-term emissions that happen over multiple days?

We do note that the majority of the VOC mass measured in these evaporation experiments was emitted within a few hours of application, however, the emissions and the timescale of emissions are also driven by temperature, film thickness, substrate material, and compound volatility. This study only sampled a few product formulations in an enclosed system at room temperature on the same substrate and did not systematically investigate temperature dependencies (Khare et al., 2020), application variability (Zhao et al., 2020), or long-term emissions (Lin and Corsi, 2007). Additionally, the PTR-ToF-MS is sensitive to many VOCs, though it is not sensitive to many larger, highly functionalized, lower volatility compounds traditionally categorized as intermediate and semi-volatile organic compounds that have long emission time-scales and have been shown to be emitted by coating products or processes (Khare and Gentner, 2018). These lower volatility compounds are likely key SOA-precursors in some regions and would need to be included in chemical transport models. While the VOCs measured by the PTR-ToF-MS could contribute to initial ozone formation following application, the changes in the emissions over time are still not well understood. The main result of this study is that VOC emissions from coatings, specifically PCBTF and Texanol, can be used as tracers for application events and usage of coatings products.

We were careful to highlight that only VOCs were measured in this study and L364-366 notes these emissions likely contribute to ozone formation, but we do not comment on any potential contributions to SOA formation. We also noted at L75-78 that SVOCs and IVOCs are key SOA precursors from coatings and have traditionally been excluded from models due to challenges in detection and quantification. We have modified the text at L364-366 to better emphasize the limitations of this study:

“For the coatings tested here, the majority of the measured VOC mass is emitted within the first few hours, and therefore the most significant atmospheric implications for ozone formation likely occur during and shortly following application, though the changes in the VOC emissions over longer timescales and the effects of environmental and application parameters were not systematically investigated in this study.”

Khare, P., Machesky, J., Soto, R., He, M., Presto, A. A., and Gentner, D. R: Asphalt-related emissions are a major missing nontraditional source of secondary organic aerosol precursors, *Science Advances*, 6(36), DOI: 10.1126/sciadv.abb9785, 2020.

Zhou, X., Gao, Z., Wang, X., and Wang, F.: Mathematical model for characterizing the full process of volatile organic compound emissions from paint film coating on porous substrates, *Building and Environment*, 182, doi: 10.1016/j.buildenv.2020.107062, 2020.