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Apportioning aldehydes: Quantifying industrial sources of carbonyls

Sarah A. Styler

Leibniz Institute for Tropospheric Research, Leipzig, Germany. E-mail: styler@tropos.de.

Photo by Ming Wang, State Joint Key Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, China.

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In their recent *Journal of Environmental Sciences* publication, Wang and colleagues provide field evidence that industrial activities can contribute substantially to atmospheric carbonyl concentrations (Wang et al., 2015). These results may help to explain underestimations of carbonyl emissions in currently available emission inventories, and highlight the need for an improved understanding of industrial sources of this class of compounds.

In the atmosphere, carbonyl compounds photolyze to yield reactive radicals, and thus contribute to the formation of ozone (Edwards et al., 2014) and other components of photochemical smog, including the NO_x reservoir peroxyacetyl nitrate (Fischer et al., 2014). Carbonyls are formed in the atmosphere as stable intermediates in the photooxidation of alkanes and other precursor species, but can also be emitted directly by a variety of anthropogenic sources, including traffic, coal burning, and industrial activities (Chen et al., 2014). An understanding of the magnitude of these primary sources is important, in part because ozone production from the photolysis of accumulated

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directly emitted carbonyls—formaldehyde in particular—would be expected to occur earlier in the day than that from carbonyls produced *via* photochemistry (Parrish et al., 2012).

The quantification of volatile organic compound (VOC) emissions is often accomplished using emission inventories, which sum the products of activity rates (*e.g.* material production) and emission factors (*e.g.* mass emission per production unit) for all known sources of a single compound or compound class (Zhang et al., 2009). Field-derived estimates of carbonyl emissions often differ substantially from those predicted by such inventories, however: in one recent study, for example, aldehyde and ketone emissions estimated from ambient measurements of these species in Beijing were found to be more than twice as high as those reported in current inventories (Wang et al., 2014). These discrepancies highlight major gaps in our understanding of carbonyl sources.

In an effort to address these uncertainties, Wang et al. measured concentrations of 72 VOCs, including aliphatic and aromatic hydrocarbons, carbonyls, and alkyl nitrates, at a rural site located in the Yangtze River Delta, China, near a number of ship manufacturing facilities. They found that VOC concentrations varied substantially over the month-long sampling campaign, with the highest concentrations—a seven-fold enhancement in total VOCs relative to clean conditions—measured during a three-day stagnant period.

This pollution period was characterized by a doubling of the relative contribution of aromatic compounds to the measured VOC profile, and in particular by substantially elevated toluene concentrations. In order to determine the source of these elevated concentrations, Wang et al. first used measured concentrations of 2-butyl nitrate and its parent alkane 2-butane to assess the extent of photochemical processing (*i.e.* the "photochemical age") of sampled air masses. Then, they used this parameter to derive the ratio of toluene to benzene in fresh air masses (*i.e.* the emission ratio) from concentrations of these species measured at the sampling site. Since benzene is primarily associated with combustion processes, whereas toluene is also emitted by industrial sources, this ratio can be used to evaluate the relative contributions of these sources to ambient toluene levels. The ratio that they obtained was significantly higher than those associated with combustion sources, which suggests that the elevated toluene concentrations measured during the pollution period were of industrial origin.

Source apportionment of carbonyls is complicated by the fact that they have both primary and secondary sources. However, since carbonyls and alkyl nitrates both arise from peroxy radical (RO_2) precursors, relationships between these two compound classes can be used to assess the relative contribution of secondary (*i.e.* photochemical) production to measured carbonyl concentrations. Although Wang et al. found a significant correlation between carbonyl–alkyl nitrate pairs under clean conditions, this correlation broke down at elevated toluene concentrations—or, in other words, when air quality at the measurement site was strongly influenced by industrial emissions. These results imply that both industrial and photochemical sources contributed to carbonyl concentrations at the site.

Extracting meaningful quantitative information regarding emissions of secondary species from correlations measured in the field is challenging (Parrish et al., 2012). Nevertheless, in order to obtain a preliminary estimate of the relative contribution of industrial activities to ambient carbonyls at the study site, Wang et al. fit measured carbonyl concentrations using a simple multiple linear regression model, in which toluene and alkyl nitrates were used as respective tracers of industrial and photochemical carbonyl sources. Using the coefficients obtained by the model, the authors estimated that industrial sources contributed ~70% of ambient acetaldehyde, ~50% of ambient acetone, and ~20% of ambient formaldehyde.

These results underscore the valuable role that field measurements can play in identifying sources underrepresented in emission inventories, and highlight the need for an improved understanding of both the magnitude of industrial carbonyl emissions and the specific industrial processes that lead to these emissions. Although not measured here, one carbonyl particularly deserving of further study is benzaldehyde, a highly efficient precursor of secondary organic aerosol (SOA) whose emissions are significantly underestimated in available inventories (Borbon et al., 2013).

Carbonyl photolysis has very recently been shown to be the dominant radical source driving ozone production in the Uintah Basin, a remote oil and gas-producing region in Utah (Edwards et al., 2014). The results obtained in the present study suggest that primary carbonyl emissions from industrial facilities have the potential to contribute to ozone production in nearby rural areas. Further work is needed, however, to clarify the contribution of these direct emissions to regional-scale air quality. Results from a recent study in Houston, Texas suggest that although primary formaldehyde emissions from petrochemical facilities are important on a local scale, regional formaldehyde concentrations are largely determined by secondary production from co-emitted reactive VOCs (Johansson et al., 2014). It is unclear whether these results are applicable to rural sites where air quality is influenced by industrial activities that do not produce large quantities of carbonyl precursors. The performance of source apportionment studies at varying distances from industrial facilities with different VOC emission profiles should help to resolve these uncertainties.

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