Carbon Tetrachloride Emissions from the U.S. During 2008 – 2012

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Carbon tetrachloride (CCl_4) is a potent greenhouse gas and an ozone depleting substance. A 100% phase-out in CCl_4 production and consumption was implemented in the developed countries in 1996 and developing countries in 2010 due to the Montreal Protocol. Only production for non-emissive uses is still allowed, such as a feedstock or process agent, and in laboratory and analytical uses. One mystery that has persisted for more than a decade is why the global atmospheric CCl_4 mole fraction is declining slower than expected based on its atmospheric lifetime and estimated emission from the reported production data; are there unidentified sources of CCl_4? If so, what do we know about these sources and their distribution? In the U.S., the U.S. Environmental Protection Agency (EPA) has been reporting zero emission since 1996. In the meantime, a few "top-down" studies also have reported approximately zero (0 – 0.5 Gg/y) emission of CCl_4 based on observations from short-term, localized-regional aircraft campaigns or two aircraft sites in the U.S. northeast and a correlation with a combustion tracer (i.e. CO or $^{14}CO_2$). However, atmospheric CCl_4 data from the U.S. portion of our Global Greenhouse Gas Reference Network suggest enhanced mole fractions at the surface relative to the free troposphere during 2008 - 2012 (Figure 1). This raises challenges to the previous findings. In this study, we will discuss the temporal and spatial variability of the surface enhancements of CCl_4 observed in our U.S. sampling network, possible sources for CCl_4 surface emissions, and the magnitude of total U.S. emissions of CCl_4 from the U.S. during 2008 – 2012 derived from inverse modeling. We will also present additional evidence from aircraft campaigns (e.g. TEXAQS) during which substantial CCl_4 enhancements were observed over the area where we infer relatively large emissions.

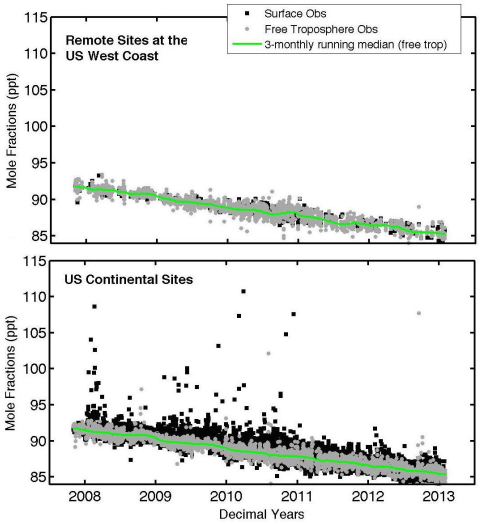

Figure 1. Observed atmospheric CCl_4 mole fractions in the free troposphere (grey) and the surface (< 1km agl) (black) at remote sites in the U.S. west coast (upper panel) and sites with more anthropogenic influence in the continental U.S. (lower panel).