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Probing the operational temperatures of vehicular catalytic converters using clumped isotopes in exhaust CO_2

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Vehicular catalytic converts are used to regulate, reduce, and convert toxic gases and environmentally harmful pollutants in exhaust gas into inert/less harmful compounds. The efficiency, however, is largely affected by the temperature of the converters. One of the major gases released is CO_2 that suffers significant re-processing in the catalytic converter; the latter process depends on the temperature of the converter. A recently utilized multiply substituted isotopologue in CO_2 namely, $^{13}C^{16}O^{18}O$, called clumped isotopes and expressed as Δ_{47}, provides a new way to probe directly the temperature of the converter. The abundance of this multiply substituted isotopologue is sensitive to the temperature at which the CO_2 is produced and/or modified. Δ_{47} is the deviation of the abundance of mass 47 in a sample CO_2 compared to a reference CO_2 in which isotopes are randomly redistributed. Here, we report Δ_{47} values of exhausts from various motor vehicles including sedans, trucks, buses, and motorcycles. In addition, for a practical assessment of catalytic converter, we sampled air CO_2 from a $\sim$13 km long highway tunnel in Taiwan inside which the CO_2 abundance is primarily controlled by the vehicle exhaust and ambient background CO_2. The average Δ_{47}values for the vehicle exhausts are 0.187$\pm$0.061 ‰ 0.077$\pm$0.027 ‰ 0.094$\pm$0.083 ‰ and 0.080$\pm$0.052 ‰ for car, bus, truck and motorcycle giving corresponding temperatures of 375, 653, 668 and 666 ^{o}C, respectively. The cars and motorcycles are gasoline operated while bus and most of the trucks are diesel operated. The Δ_{47}values indicate that the catalytic converters are operated at lower temperatures for the cars compared to bus, truck and motorcycles. The Δ_{47}values of tunnel CO_2 range between 0.308 and 0.718 ‰ at [CO_2] of 3326 and 646 ppmv, respectively corresponding to effective temperatures of 250 and 71 ^{o}C. The end-member Δ_{47}value for the tunnel CO_2, derived from a two-component mixing model [1], was found to be 0.141$\pm$0.026 ‰ giving a temperature of CO_2formation of 450 ^{o}C which is similar to the average value of the vehicle exhausts. Temperatures inside vehicle's combustion chambers are much higher ($\sim$800 ^{o}C for gasoline engine and even higher for diesel engines) than that estimated by the Δ_{47} values. It is observed that the temperature of catalytic converter depends on the vehicle speeds and decomposition/oxidation of harmful chemicals such as CO and NO_x depends on the temperature of catalytic converter. Clumped isotopes in exhaust CO_2 can be a potential tracer to assess the efficiency of vehicular catalytic converters at diverse operational conditions. The results may also have wide implications in estimating the anthropogenic contribution of CO_2 using bottom up approach in urban areas.

References

1 **Laskar, A. H., et al. (2016). Identification of anthropogenic CO_2 using triple oxygen and clumped isotopes. Environ. Sci. Tech. 50 (21), 11806–11814.**