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A numerical study on elevated levels of HCHO within ship-plumes: Source identification and budget analysis

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Elevated levels of formaldehyde (HCHO) along the ship corridors have been observed by satellite sensors, such as ESA/ERS-2 GOME (Global Ozone Monitoring Experiment), and were also simulated by global 3D chemistry-transport models. The elevated HCHO levels along the heavy ship-traffic corridors are important because they can greatly perturb atmospheric oxidation cycle in the ship-influenced marine boundary layer (MBL). In this study, three likely sources of the elevated HCHO levels in the ship plumes as well as their contributions to the elevated HCHO levels were investigated using a ship-plume photochemical/dynamic model: (i) primary HCHO emission from ships; (ii) secondary HCHO production via the atmospheric non-methane volatile organic compounds (NMVOCs) emitted from ships; and (iii) atmospheric oxidation of CH_4within the ship plumes. For this numerical analysis, the ITCT 2K2 (Intercontinental Transport and Chemical Transformation 2002) ship-plume experiment was chosen as a base study case. From multiple model simulations for this base case, it was found that main responsible factor for the elevated HCHO levels is CH_4 oxidation by elevated levels of in-plume OH radicals. More than $\sim$88% of the HCHO for the ITCT 2K2 ship-plume is produced by this atmospheric chemical process, except in the areas close to the ship stacks where the main source of the elevated HCHO levels would be primary HCHO from the ships. Because of active CH_4oxidation by OH radicals, the instantaneous chemical lifetime of $CH_4(\tau_{CH4})$ decreased to $\sim$0.45 yr inside the ship plume, which is in contrast to τ_{CH4} of $\sim$1.1 yr in the background (up to $\sim$41% decrease) for the ITCT 2K2 ship-plume case. A variety of likely ship-plume situations at three different latitudinal locations within the global ship corridors was also studied to determine the enhancements in the HCHO levels in the MBL influenced by ship emissions. It was found that the ship-plume HCHO levels could be 19.9-424.9 pptv higher than the background HCHO levels depending on the latitudinal locations of the ship plumes, MBL stability and NO_x emission rates. On the other hand, NMVOC emissions from ships were not found to be a primary source of photochemical HCHO production inside ship plumes due to their rapid and individual dilution.