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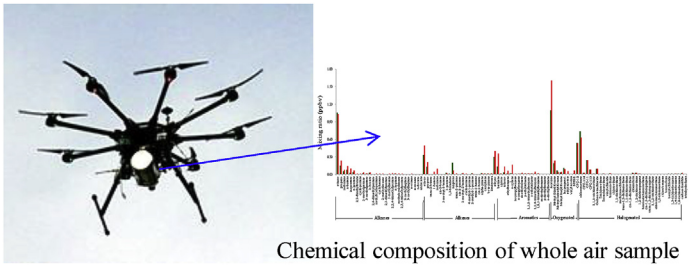
A study of atmospheric mixing of trace gases by aerial sampling with a multi-rotor drone

Chih-Chung Chang[a,*], Chih-Yuan Chang[a], Jia-Lin Wang[b], Ming-Ren Lin[a], Chang-Feng Ou-Yang[b,c], Hsiang-Hsu Pan[a], Yen-Chen Chen[a]

[a] Research Center for Environmental Changes, Academia Sinica, Taipei 11529, Taiwan, ROC
[b] Department of Chemistry, National Central University, Chungli 320, Taiwan, ROC
[c] Department of Atmospheric Sciences, National Central University, Chungli 320, Taiwan, ROC

GRAPHICAL ABSTRACT



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ABSTRACT

We exploited a novel sampling vehicle, a multi-rotor drone carrying a remote-controlled whole air sampling device, to collect aerial samples with high sample integrity and preservation conditions. An array of 106 volatile organic compounds (VOCs), CO, CH_4, and CO_2 were analyzed and compared between the aerial samples (300-m height) and the ground-level samples in pairs to inspect for vertical mixing of the trace gases at a coastal site under three different meteorological conditions of local circulation, frontal passage, and high-pressure peripheral circulation. A rather homogeneous composition was observed for the sample pairs immediately after the frontal passage, indicating a well-mixed condition below 300 m. In contrast, inhomogeneous mixing was observed for the sample pairs under the other two conditions (local circulation and high-pressure peripheral circulation), suggesting different layers of air masses. Furthermore, information of unique source markers, composition profiles, and lifetimes of compounds were used to differentiate the origins of the air masses aloft and at the surface to substantiate the observed inhomogeneity. The study demonstrates that, with the availability of the near-surface aerial sampling coupling with in-laboratory analysis, detailed compositions of trace gases can now be readily obtained with superior data quality. Based on the distinctive chemical compositions, the sources, transport, and atmospheric mixing of the airborne pollutants in the near-surface atmosphere can be better studied and understood.

1. Introduction

Trace gases (e.g., volatile organic compounds (VOCs), CO, CH_4, and CO_2) play crucial roles in many important environmental issues, involving atmospheric secondary oxidant formation (e.g., organic peroxy radicals (RO_2), ozone (O_3), and secondary organic aerosols (SOA)),

* Corresponding author.
E-mail address: joechang@rcec.sinica.edu.tw (C.-C. Chang).

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tropospheric air pollution, stratospheric ozone depletion, regional and global climate, as well as toxicity concerns (Atkinson and Arey, 2003; Blake and Rowland, 1995; Hallquist et al., 2009; Molina and Rowland, 1974; IPCC, 2013). These trace-gas related issues usually mainly or partially take place within the planetary boundary layer (PBL) where emissions and atmospheric processing of trace gases occur most vigorously (Garratt, 1994). To gain insightful knowledge of the mixing, transport, and dispersion of trace gases, as well as their relations with meteorology in the boundary layer, speciated analyses to obtain detailed composition of trace gases is basic and essential (Velasco et al., 2008; Aurell and Gullett, 2013; Ashfold et al., 2015; Greenberg et al., 1999; Chen et al., 2002; Glaser et al., 2003; Corrigan et al., 2008). In the past, such knowledge was often obtained via sampling or measurements at the surface, which is susceptible to over-influence by sources in the proximity and; therefore, the lack of area representativeness could easily lead to bias in source investigation. Although aerial observations on trace constituents at other altitudes above ground may suggest otherwise, technical difficulties, lack of proper tools and high cost often limit the availability and implementation of the near-surface airborne measurements.

Recently, the technique of taking low-altitude aerial samples using drones has matured (Chang et al., 2016). A multicopter carrying sampling flasks aloft can collect whole air samples at the desired height while hovering. Compared to other means of elevated sampling methods such as high towers, tethered balloons, and fixed-wing aircrafts, sampling with multicopter drones has many obvious advantages. Drones' unique ability to hover at a fixed location, move vertically and maneuver with agility make aerial sampling an effective way to obtain the vertical composition of airborne trace gases in the lower atmosphere. A better representation of airborne trace gases' composition and levels can shed light on the emission sources, loss processes and mixing conditions during transport.

Volatile organic compounds (VOCs) are a principal group of trace constituents in the atmosphere, originating from a variety of sources (e.g., burning of coal and oil, industrial emissions, solvent evaporation, biomass burning, and other anthropogenic and biogenic emissions). Each type of sources usually bears its own characteristic composition of VOCs (Guo et al., 2006; Chang et al., 2009; Liu et al., 2012). Furthermore, VOC species exhibit considerable differences in chemical and physical properties (i.e., reactivity, polarity, solubility, volatility). The chemical and physical sinks together determine their atmospheric residence times (or lifetimes) (Atkinson and Arey, 2003). The observed composition of VOCs can be linked to possible sources, and a range of species of various lifetimes can be used to infer distances or the age of an air mass from emission sources, at least in a relative sense (Gelencsér et al., 1997; Ashfold et al., 2015; Miller et al., 2011).

In this study, aerial and ground-level whole air samples were collected simultaneously in pairs at a coastal headland site in various types of meteorological conditions. The aerial samples were taken by a multi-rotor platform carrying a whole air sampling device. An array of 106 VOCs, covering a wide range of sources and atmospheric reactivities, as well as CO, CH_4, and CO_2 were analyzed to examine atmospheric mixing of trace gases in the near-surface atmosphere and to infer the possible sources and transport pathways of air masses with specific composition under different meteorological conditions.

2. Experimental

2.1. Geographic domain and meteorology

The launch site of aerial sampling is at the northern tip of Taiwan (Cape Fuguei), as shown in Fig. 1. Cape Fuguei's geographic location, surrounded by ocean on three sides, makes this headland an ideal location to receive airborne pollutants from East Asia, the Asian continent, the northwest Pacific Ocean and the island itself. Field experiment was conducted in autumn. In the season, the Asian continental

Fig. 1. Sampling site at Cape Fuguei in northern Taiwan in East Asia.

cold high-pressure system strengthens, and the weather pattern transitions from typical summer conditions with warm and mild local circulation to the cold surge-dominated conditions that manifest as colder temperatures and northeasterly winds (Chang et al., 2006b). In addition to these two major weather conditions (local circulation and cold surge), a transitional meteorological condition (high-pressure peripheral circulation) usually occurs after a stronger cold surge. When the continental high-pressure system moves from China to the West Pacific, its peripheral circulation blows clockwise to deliver a mild southeasterly/easterly wind around the island of Taiwan, and the weather becomes warmer with a mild breeze.

During the period of the field experiment, from 27 October to 6 November 2015, one weaker front (27–28 October) and one stronger front (27 October-3 November) passed through Cape Fuguei. Fig. 2 shows warm conditions with a mild breeze (0–5 m/s) prior to the cold fronts. As the fronts arrived, the temperature dropped, and the winds swiftly changed to stronger northeasterlies (5–12 m/s). After the stronger cold surge (3 November), the weather pattern transitioned to high-pressure peripheral circulation (3–6 November).

2.2. Sampling and analysis of trace gases

Aerial and ground-level samples were taken simultaneously in pairs at the coastal site under the above-mentioned three meteorological conditions (local circulation, frontal passage, and high-pressure peripheral circulation). The surface samples were taken manually overhead at approximately 2 m above the surface, and the aerial samples were taken by the multi-rotor platform at a height of 300 m. Totally eleven pairs of samples were collected in the three different meteorological conditions, i.e., four pairs for local circulation (in blue), three pairs for frontal passage (in red), and four pairs for high-pressure peripheral circulation (in green), as indicated by the colored bars in Fig. 2. During the mission period, we needed to seize the opportunity to collect samples at the right moment in the three different meteorological conditions, resulting in irregular sampling time and quantities. The exact sampling time of each sample pair during each meteorological