Atmospheric Environment 84 (2014) 78–87

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Enhancement of the hygroscopicity parameter kappa of rural aerosols in northern Taiwan by anthropogenic emissions

Hui-Ming Hung [a,*], Wan-Jung Lu [a], Wei-Nai Chen [b], Chih-Chung Chang [b], Charles C.-K. Chou [b], Po-Hsiung Lin [a]

[a] Department of Atmospheric Sciences, National Taiwan University, No. 1, Sec. 4, Roosevelt Road, 10617 Taipei, Taiwan
[b] Research Center for Environmental Changes, Academia Sinica, Taipei, Taiwan

HIGHLIGHTS

- The refractive indices without absorption part are 1.40–1.52 for rural site.
- The nucleation event at rural site is more obvious with anthropogenic emission.
- The aerosol hygroscopicity at rural sites is higher than urban regions in Taiwan.
- The hygroscopicity of rural aerosols is enhanced by the anthropogenic emission.
- The derived κ is size dependent, ranged from 0.05 to 0.35.

ARTICLE INFO

Article history:
Received 2 July 2013
Received in revised form
6 September 2013
Accepted 13 November 2013

Keywords:
Aerosol
Cloud
CCN
Hygroscopicity
Refractive index

ABSTRACT

The hygroscopicity of aerosols in a rural site of northern Taiwan, HuaLin, for August 2011 was derived to investigate the impact of anthropogenic emission on the physico-chemical property of aerosols using the measured particle number size distribution, condensation nuclei (CN) concentration and the number of particles activated at supersaturation (SS) of 0.089–0.91%. Based on the concentration of anthropogenic volatile organic compounds (VOCs) and the back trajectory information, this campaign was divided into two periods: with and without significant anthropogenic contributions as 8/12–8/19/2011(Period_1) and 8/19–8/22/2011(Period_2), respectively. The refractive indices without absorption calculated using the measured scattering coefficients and number size distribution of aerosol particles were 1.43 ± 0.03 for 8/13–8/17 and 1.49 ± 0.03 for 8/18–8/21, having a consistent trend with back trajectory and the selected VOC concentrations. The new particles formation happened more significant for Period_1 and likely to be associated with the higher concentration of anthropogenic emission such as SO_2 or VOCs which went through the oxidation or photo-oxidation to form more condensable species for nucleation. The derived single hygroscopicity parameter increases with particle size and was higher for Period_1 (0.1 at 50 nm– 0.35 at 165 nm) than Period_2 (0.04 at 70 nm– 0.28 at 175 nm) possibly due to the addition of aged composition as the air parcel passed through cities and went through photo-oxidation processes before arriving HuaLin. However, the hygroscopicity of rural aerosols tends to be higher than the urban aerosol in this study possibly due to the aging level. This study suggests the important role of anthropogenic emission on the secondary aerosol formation and also the physical properties of aerosols over rural area.

1. Introduction

Aerosol particles play an important role affecting climate such as reflecting or absorbing radiation directly (IPCC, 2007; Seinfeld and Pandis, 2006) and acting as cloud condensation nuclei (CCN), which influence the cloud albedo and also the hydrological cycle (Facchini et al., 1999; Lohmann and Feichter, 2005; Murphy, 2005; Tunved et al., 2006; Twomey, 1974, 1977). The efficacy of aerosol particles becoming CCN is strongly dependent on the physico-chemical properties of the particles (Martin, 2000). If the aerosol particles are composed with high hygroscopic species such as sulfate, nitrate or oxalic acid, the aerosol particles would take up significant of water when the relative humidity (RH) is high enough and further grow into cloud droplets at high enough water supersaturation conditions (Seinfeld and Pandis, 2006). The cloud droplets formed at high CCN concentration would be

* Corresponding author.
E-mail address: hmhung@ntu.edu.tw (H.-M. Hung).

1352-2310/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.atmosenv.2013.11.032

smaller than that at low CCN concentration. The size and number concentration of cloud droplets would further provide different scattering efficiency on the solar radiation and also affect the strength and quantity of precipitation. The influence of cloud on climate exits a significant uncertainty according to the IPCC report (2007) due to the complex factors such as aerosol composition, number-size distribution and lifetime. In addition to the strong influence on radiation, cloud droplets can also affect the atmospheric chemical reactions such as the oxidation of SO_2 and hydrolysis of N_2O_5 due to the significant larger moisture surface area as compared with the haze particles (Broekhuizen et al., 2004; Cassar et al., 2007; Meskhidze and Nenes, 2006; Robinson et al., 2007; Rudich et al., 2007).

For a well known composition of aerosol particles, the size variation as a function of RH can be estimated using Köhler equation which includes the Raoult's law and Kelvin effect (Seinfeld and Pandis, 2006). However, the ambient aerosol composition is a complex, usually a mixture of various species such as inorganic, organic, soluble and insoluble species contributed directly or indirectly by anthropogenic and natural sources. Due to the oxidative atmospheric environment, there are new particle formation and possible aerosol composition variation with time via the aging processes such as photolysis or oxidation with ozone or OH radicals (Burkart et al., 2011; Rose et al., 2011, 2010; Yue et al., 2011). The CCN activity of aerosol particles then has time characteristics in addition to the geographic distribution (Burkart et al., 2011; Gunthe et al., 2011; Kondo et al., 2010; Kumar et al., 2003; Kuwata et al., 2009; Rose et al., 2011, 2010; Yue et al., 2011). To quantify the influence of ambient aerosol particles on cloud formation, the CCN activity of aerosol particles has been extensively studied in laboratories and field measurements (Abbatt et al., 2005; Corrigan and Novakov, 1999; Gunthe et al., 2011; Kondo et al., 2010; Kumar et al., 2003; Kuwata et al., 2009). Thus the ability of aerosol particles acting as CCN for different environments is required for further investigating the impact of anthropogenic emissions on the global climate change.

Taiwan is an island in the eastern Asian Pacific Ocean with an area of 35,883 km^2. The Central Mountain Range runs from the north to the south of Taiwan as the principal range of mountains in Taiwan. The major metropolitan cities are mainly located in the west of Taiwan and act as the major local anthropogenic emissions in Taiwan. The geographical distribution in Taiwan indicates the significant interaction between biogenic and human activities. The air quality of Taiwan is affected from regional transportation based on the meteorology conditions such as the Asian outflows during winter and transportation from southwest during summer, in addition to the local emission. For urban sites, the air quality is mainly controlled by the ventilation of the local emission in addition to the regional transportation. In rural region, the secondary organic aerosol formation from the photooxidation of biogenic emission may play an important role to affect the overall physico-chemical properties of the ambient aerosols (Levin et al., 2012; Poeschl et al., 2010). However the influence of the anthropogenic emission through short-range regional transportation might modify the physical properties of aerosols in rural sites.

In this study, we report a detail characterization of the hygroscopicity of rural aerosols in northern Taiwan for 2011 summer. The aerosol diameters required to be activated into cloud droplets were determined for two different levels of anthropogenic contribution defined by the back trajectory and the concentration of volatile organic compounds (VOCs). The physical properties of aerosol particles such as optical constants and single hygroscopicity parameters were derived to investigate the influence of anthropogenic emission.

2. Experimental methods

The measurements were conducted over a rural site of northern Taiwan, HuaLin, (25.89°N, 121.57°E with 410 m of altitude), which is approximate 15 km away from the Taipei city and 10 km away from the closest city, Sindian. There is a Taiwan EPA station in Sindian to provide the concentrations of CO, NO_x, SO_2 and O_3. The satellite image of northern Taiwan with the indication of HuaLin and National Taiwan University (NTU) as an urban site is shown in Fig. 1. The environment of HuaLin is surrounded mainly with bushes and trees and the road traffic is limited. The campaign was conducted from 07/04/2011–08/22/2011 (LT = GMT + 8:00). Due to the limited available instrumentation and complete dataset for the required analysis, this study focuses on the time period of 08/12/2011–08/22/2011. The study over NTU site was conducted in the period of June 6–15, 2012 and limited to the measurement of CCN activity for comparison.

2.1. Instrumentation

The meteorological parameters such as temperature, RH, wind direction and wind speed were monitored using an All-In-One compact weather sensor (Climatronics Corp. USA). The radiation was detected with a Li-190 Quantum Sensor (NexSens Technology, USA) for the photosynthetically active radiation. The concentration of VOCs was determined using a gas chromatography (Varian CP-3800) with parallel detectors (Varian Saturn 2200 ion-trap mass spectrometer and flame ionization detector, FID) for the collected gas sampled with canisters (Chang et al., 2010a; Wang et al., 2012). Other instruments used for the physico-chemical properties of aerosols are illustrated in detail as follows.

2.1.1. Cloud activation measurement

The schematic setup for the CCN activation measurement is shown in Fig. 2 with three major systems; a cloud condensation nuclei counter (CCNc), a condensation particle counter (CPC) and a scanning mobility particle sizer (SMPS). The CCN measurements were conducted using a thermal-gradient CCNc (Droplet Measurement Technologies, Colorado) (Roberts and Nenes, 2005) while the total number of condensation nuclei were monitored using a CPC (TSI 3776). The ambient aerosols were characterized by a SMPS (TSI 3936) composed with an Electrostatic Classifier (TSI 3081) and a CPC (TSI 3772) to obtain the number-size distribution at size range of 9–430 nm and a sampling cycle of 5 min. The sampling flow of 1.1 L min^{-1} passed through a 1 m long of diffusion dryer filled with silica gel to reduce the RH to less than 35% before entering CCNc, CPC and SMPS in parallel. The temperature gradient (ΔT) of CCNc were varied in the range of 3–16 K, corresponding to supersaturation (SS) of 0.089–0.91% based on the SS calibration using ammonium sulfate particles with the osmotic coefficient as a function of ammonium sulfate concentration (Ally et al., 2001). For each given ΔT, the determined critical diameter can be converted to the calibrated SS values as summarized in the Supplementary data. Five SS conditions were set with a duration time of 10 min for each SS. However, the first few points were removed in the moment of SS switched due to the transient process to reach the new temperature gradient.

2.1.2. Scattering coefficient

A NCN-2A (Optec Inc.) nephelometer composed with a heating set was applied to measure the aerosol scattering coefficients at dehydration condition. The wavelength for the nephelometer is 550 nm with integration angle from 5° to 175°. The nephelometer was regularly calibrated using zero air or calibration gas (R13a) to insure the performance through the whole campaign. The