FLUORESCENCE DATABASE OF AEROSOL-CANDIDATE-SUBSTANCES FOR FLUORESCENCE LIDAR APPLICATION

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ABSTRACT

A database containing spectrum and cross-section of the fluorescence of substances has been made. In test of forest environment monitoring by our Laser-Induced Fluorescence Spectrum (LIFS) lidar, the database showed that the origin substance of the aerosol observed by the lidar was cedar pollen, and the concentration was calculated using the cross-section. In the urban atmosphere monitoring, three substances stored in the database were proposed to be the origins of the aerosol. Based on these experiments, we discuss the usefulness of the fluorescence database in lidar observations.

1. INTRODUCTION

Until recently, using fluorescence lidars had been limited to hard targets such as vegetation, water, living things, and buildings because low cross-section of aerosol fluorescence makes dear observations difficult. However recent spectrum detection technologies using an intensified CCD or a multi anode photomultiplier single-photon-counting have been expanding the fluorescence lidar observation capabilities to atmospheric aerosol monitoring where the target concentrations are much less than that of hard targets.

Tatarov et a.l(1) and Saito et al.(2) first reported detections of the fluorescence spectrum from the atmosphere in KOSA events at the same time but independently using their own different fluorescence lidars. Sugimoto et al. showed that fluorescence in the lower atmosphere could be of air-pollution aerosols transported from urban and industrial areas(3). The co-existence of clouds and fluorescing aerosols was discussed by Reichadt et al.(4)

The problem in lidar atmospheric observations is how to identify the species of aerosol observed by the lidar. Even if we use a fluorescence lidar, the fluorescence information of substances is needed.

In this paper, we describe a fluorescence database developed for this purpose. It contains fluorescence information about the spectrum and cross-section of substances which we can use in the discussion of fluorescence lidar observations.

2. FLUORESCENCE LIDAR OBSERVATIONS

Two types of LIFS lidars were developed (Fig.1). Both systems use a 355-nm laser for the lidar transmitter and a multichannel spectrometer for the lidar spectroscopic receiver.


(a) (b)

Figure 1. (a) LIFS-field lidar and (b) LIFS-lab lidar.

2.1 Pollen fluorescence monitoring by LIFS-field lidar(5)

Many people suffer from pollen allergies, so we decided to test the possibility of pollen monitoring by a fluorescence lidar.

A fluorescence lidar capable of operating outdoors was developed at Shinshu University and named the LIFS-field lidar. We used a multi-channel spectrometer (PMA-12; Hamamatsu, Japan) for spectral detection which consisted of a spectrometer with a gated-intensified CCD array detector. Its short gating time of several tens of ns made it possible to detect weak fluorescence

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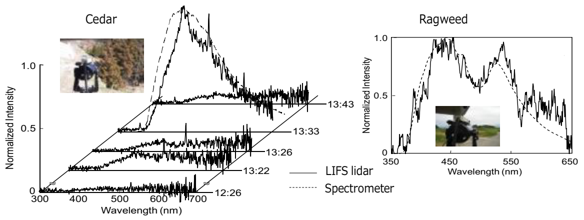
signals even in daytime. The LIFS-field lidar was a mobile and self-sufficient system operating with a small dynamo (900 VI).

Spring-time observations were made at the mountain-side of a town (Matsushiro, Nagano City, Japan) where there was a forest with several different species of trees. The lidar was placed on the ground so that it was about 3 m higher than the ground of the forest. The distance to the forest was about 20 m. The detected fluorescence spectrum is shown in Fig. 2a. The signal reached the peak at 13:33. Time depended signal intensity variation was caused by the wind.

Autumn observations were made at a riverbed of the town where the ragweed had spread over large areas. The laser beam passed horizontally 1 m above the ragweed area. Fig. 2b shows the detected fluorescence spectrum which has a different spectral shape from the cedar and the intensity was slightly lower than that of cedar pollen.



(a) (b)

Figure 2. (a) Cedar and (b) ragweed pollen monitoring by the LIFS-field lidar. Each of the spectra stored in the database are comparatively shown in the observation.

2.2 Atmospheric aerosol fluorescence monitoring by LIFS-lab lidar

Fukuoka has a population of over one million. Above the troposphere, the westerly wind constantly flows carrying KOSA particles, and the area faces the Japan Sea. Investigation of urban atmosphere is one of our research interests, since they can be affected by various interactions among the atmosphere, sea, and human activity.

A LIFS-lab lidar was developed at Fukuoka University for observing urban atmospheric environments. The detection system was consisted of a spectrometer (SP-2758, Acton, USA) and a 32-channel multi-spectral lidar detector (MS-125, Licel, Germany). The LIFS-lab lidar had functions of Raman and depolarization measurement. Multiple wavelength lidar using 532 nm and 1064 nm also worked together with the LIFS-lab lidar.

A laser beam was transmitted vertically and observation was done at night automatically and routinely. A fluorescence signal was observed in almost every observation. One of the examples is shown in Fig. 3 in the state where urban air pollution and KOSA particles were mixed. The signal intensity was saturated in the atmosphere lower than 1 km and doubled compared with only KOSA particle inflow. Many other observations showed that the fluorescence signal was higher in KOSA events and decreased if the depolarization was high, and it increased if the particles were liquid-oriented ones.

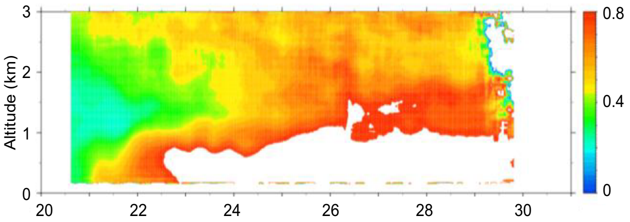


Figure 3. An example of the urban atmosphere in a KOSA event monitored by the LIFS-lab lidar. Total fluorescence signal intensity was normalized to N2 Raman signal at 420-510 nm.

3. FLUORESCENCE DATABASE

3.1 Preparation of fluorescence substance

Traditional lidar analysis seems to have mainly dealt with inorganic substances. Our scenario of aerosol generation is conceptually illustrated in Fig. 4, in which we examine every kind of

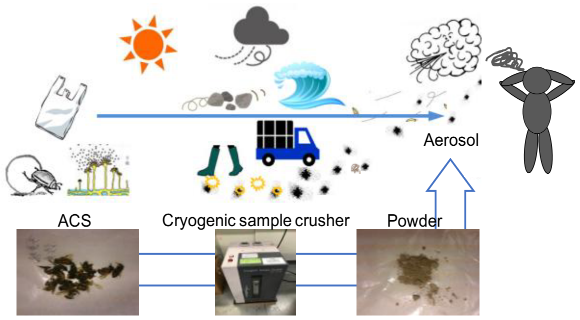


Figure 4. Scenario of aerosol generation (upper) and procedure of making powder of ACS (lower).

substance without limiting the type.

Plastic garbage, plants, living things, and minerals are included. Here we named this Aerosol-Candidate-Substance (ACS) which can be expected to be aerosols.

ACSs were collected from the surroundings and made into powder by a cryogenic sample crusher (JFC-3000, Japan Analytical Industry, Japan) to resemble aerosols in the atmosphere. The size after crushing was around 50 µm. ACSs such as pollen, which is a powder naturally (particle), was used as it is without crushing.

3.2 ACSs Fluorescence spectrum

Excitation-Emission-Matrix (EEM) characteristics of each powder were measured by a fluorescence spectrometer (F-7000, Hitachi High-Tech Science, Japan).

Several examples of EEM are shown in Fig. 5. The emission (fluorescence) spectrum was measured by varying the excitation wavelength from 250 or 300 nm to 540 nm with 10 nm separation.

We developed software which is useful in identifying aerosol species in lidar observations and installed it into the fluorescence database. The database stores fluorescence spectra of 50 species so far and they are roughly divided into plant-, biological-, and artificial-related ones.

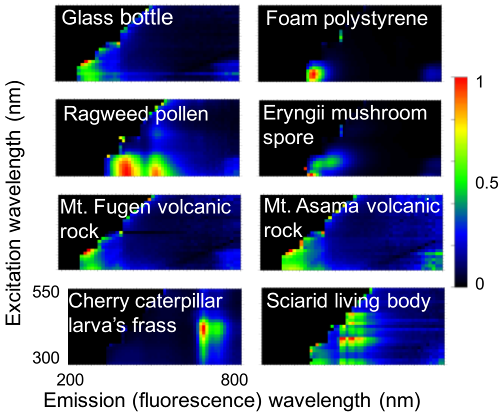


Figure 5. Examples of EEM of ACS powders.

3.3 ACSs fluorescence cross-section

Information about cross-section (or quantum efficiency) of aerosol is needed to calculate aerosol concentrations from lidar observations, but there are very few reports on the data. In this work, we developed a fluorescence cross-section measurement system. A 355-nm pulse laser irradiated ACS powder set on a non-fluorescent quartz plate and a photomultiplier measured the voltage of the fluorescence. A particle counter (KC-20, RION, Tokyo, Japan) determined the number of powdered particles of the ACS.

Cedar pollen was investigated. Figure 6 shows the fluorescence intensity (voltage) depending on the number of particles. From this we could derive that the fluorescence cross-section of cedar pollen was 1.5x10^{-13} cm^{2} sr^{-1} nm^{-1} particle^{-1} at 460 nm. This is compatible with that of pine pollen reported by Stephen$^{6)}$ and those of Betula alba and other pollens by Weichert$^{7)}$. It should be added that their excitation was at 280 nm.

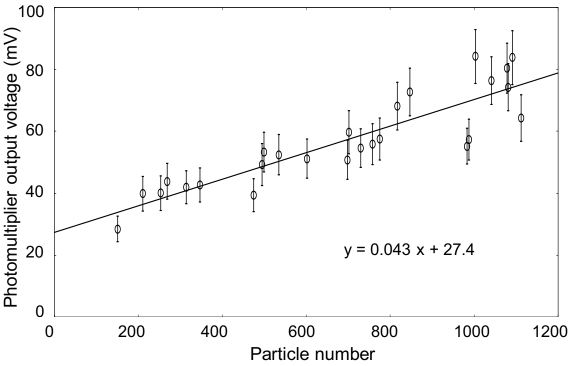


Figure 6. Fluorescence intensity of cedar pollen depending on the numbers of particles.

4. ADAPTATION OF DATABASE

4.1 Pollen observation$^{5)}$

There were several species of trees growing in the area where pollen observations were made as described in 2.1. They included locust tree, cedar, pine and cypress. Fluorescence spectrum of each stored in the database helped us identify the species, so that we knew that the spectrum observed by the LIFS-lab lidar was of cedar pollen. In the autumn observation, it was ragweed pollen.

We calculated the concentration of the cedar pollen by applying the cross-section data obtained in 3.3 in the lidar result. In this case, the sensitivity of the PMA detection of the LIFS-field lidar was checked by a comparative experiment with a photomultiplier detection. Then the calculation indicated that the number of cedar pollens observed by the LIFS lidar was 1.5x10^7 particle m^{-3}.

4.2 Atmospheric aerosol observation[8)]

We investigated ACSs which might be the origin of aerosols observed by the LIFS-lab lidar.

As the Environmental Scanning Electron Microscope Energy Dispersive X-ray (ESEM-EDX: Quanta FEG 200, FEK and XL-30, EDAX, U.S.A) analysis of the substances sampled on a building roof suggested that aerosols included mostly Si with Al, Mg, and smaller concentrations of Fe, S, Ca, K, we selected three fluorescence spectra from the database which might contain such minerals. These included the fluorescence of glass bottles, Mt. Asama volcanic rock, and concrete blocks.

Figure 7 compares the fluorescence spectra of atmospheric aerosols and a composition of three ACSs and their tendencies of increasing and decreasing were similar. Among the three ACSs, the spectrum of a glass bottle, whose main component is SiO_2, seemed to fit well.

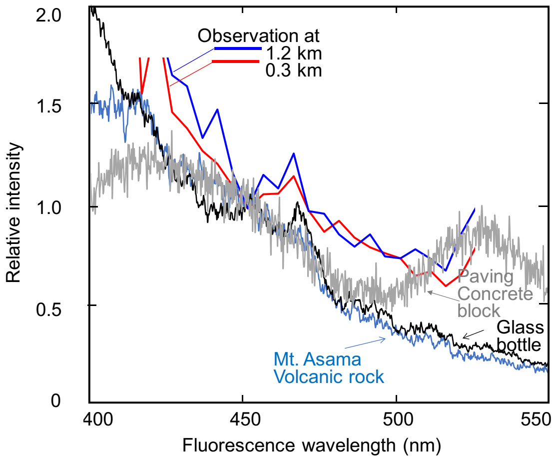


Figure 7. Adaptation of fluorescence spectrum of three ACSs into lidar atmospheric fluorescence data. The values of each spectrum were compared relatively with their own intensities at 452.4 nm.

5. CONCLUSION

We developed two types of LIFS lidars that made it possible to observe atmospheric fluorescence. Fluorescence spectrum database was useful in estimating the origins of atmospheric aerosols. Cedar pollen concentrations could be estimated by applying its cross-section value into the computation.

These experimental results show that the LIFS lidar with a fluorescence database is a new apparatus for conducting research of atmospheric environments.

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