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学位論文題目	Evaluation of a newly developed ultrafine particle sampler and its application to quantification of chemical components in atmospheric ultrafine and fine particles （新規開発超微粒子捕集装置の評価とその大気中超微小粒子と微小粒子中の化学成分の同定への応用）
論文審査委員	委員長　教　授　坂本　和彦 委　員　教　授　吉門　洋 委　員　准教授　王　青躍 委　員　連携准教授　三輪　誠

論文の内容の要旨

There are many atmospheric pollutant emissions from biogenic and anthropogenic sources both in gaseous and particulate phases. The atmospheric particulate matters can be divided into two sections, primary and secondary particulate matter. The term “primary particulate matters” is defined as the particulates directly emitted from emission sources while the secondary particulate matters is defined as the particulates newly formed via nucleation or changed their original forms via gas-to-particle conversion by photochemical reactions or oxidations. The particulates containing organic carbon (OC) mainly undergo these alterations which makes us difficult to estimate the pollutant emission sources thus the atmospheric particulates measurement with gaseous phase would provide us very useful information to tract the original emission sources. Atmospheric processing in particulates undergoes further with the air mass transportation and this makes the pollution level from local to regional, and regional to global. The particulates either can exist in the atmosphere or can be deposited on the surface in dependence on their respective sizes. The particulates which can exist in the atmosphere for a long time have a significant climate impact so called, global warming, cooling or regional cooling by the particulates containing black carbon (elemental carbon) and organic carbon. Fine particles (FPs, $D\mathrm{p} < 2.5\ \mu\mathrm{m}$) are mainly responsible for these climate impact due to the longest residence time in the atmosphere thus measurement of fine particles is crucial. Furthermore, particulates can also cause adverse health effect on human. Especially, ultrafine particles (UFPs, $D\mathrm{p} < 0.1\ \mu\mathrm{m}$) can penetrate into our lung deep inside (Alveolar regions) approximately 70%. Thus, the measurement of UFPs is also very important. Since the main removal mechanism of UFPs is a particle growth into fine particle size by coagulation, it would be very useful to measure the two particle sizes simultaneously. In recent studies, it has been shown that UFPs make the highest contribution to the total particle number concentrations, while only a small contribution to particle mass and that accumulation mode particles ($0.1 < D\mathrm{p} < 2.5\ \mu\mathrm{m}$) make the highest contribution to the total particle mass concentrations in urban environment. The ultimate objectives of this study is: 1) to provide accurate chemical

composition of ambient ultrafine and fine particles, 2) to provide temporal variation of ultrafine and fine particles, 3) to find a relationship between the two particle sizes in chemical composition, and 4) to estimate the emission sources qualitatively. In order to achieve these objectives, first of all, the sampling method for UFPs has to be considered. Due to the difficulties in the analysis of chemical composition in ultrafine particles with the existing sampling method, a newly developed sampling method (an INF sampler) was applied in this study.

The INF sampler enables us to collect sufficient UFPs mass for relatively short time interval, say 24 h, which has been considered almost impossible with the existing UFPs samplers. These series of studies indicate several findings: 1) 24h or even 12h time interval UFPs mass collection in the ambient air might be feasible with this INF sampler, 2) It became feasible to observe the chemical composition of ultrafine and fine particles simultaneously which has been considered almost impossible for the existing samplers, 3) The simultaneous UFPs and FPs collection enables us to tract the fundamental emission sources and the contribution of UFPs to FPs in the atmosphere, 4) The characteristics of UFPs in comparison with FPs in chemical composition can be observed, 5) Using the annular denuder technique, simultaneous collection of gaseous and particulate phase in ultrafine particles can be achieved providing us unbiased particle collection with the information of gas-to-particle conversion in UFPs. This study is very valuable as the first study which achieved 24h time interval UFPs sampling with simultaneous particle collection of FPs and found the relationship between UFPs and FPs in chemical composition with characteristics and possible emission sources.

論文の審査結果の要旨

当学位論文審査委員会は、当該論文の発表会を平成 22 年 8 月 9 日に公開で開催し、約 40 分の発表の後、本論文に関する詳細な質疑を行い、論文内容を審査した。

以下に論文内容を示し、学位論文審査の結果を要約する。

Chater 1 General introduction

The importance of particulate matter (PM) in ambient air with the chemical and physical properties is introduced especially for ultrafine particles (UFPs) in this Section. Also, toxicological concerns with the environmental regulations for PM in other countries are addressed. To measure PM especially for UFPs, it is very crucial to know particle collection techniques and analytical methods thus collection techniques with analytical methods and sampling strategies for UFPs collection are introduced. The objectives of doctorial thesis are clarified at the end of this Chapter.

Chapter 2 Performance test of an inertial fibrous filter for ultrafine particle collection and the possible sulfate loss when using an aluminum substrate with ultrasonic extraction of ionic compounds

Recently developed UFPs sampler (INF sampler) was tested for generated particles in the laboratory and for ambient particles in the atmosphere compared with a nano-MOUDI sampler which is world widely used for the collection of nano, and ultrafine particle. The critical point of this Section is that the possible sulfate loss when using an aluminum substrate with ultrasonic extraction of ionic compounds is addressed by analyzing aluminum ions in the extracted solution with ICP-MS. Overall, the INF sampler showed excellent UFPs collection in terms of chemical composition comparison between INF and nano-MOUDI sampler.

Chapter 3 Seasonal variation of carbonaceous, ionic components in fine and ultrafine particles

As an application of INF sampler for UFPs collection, seasonal variation of carbonaceous, ionic components in fine and ultrafine particles was studied. Furthermore, the information of chemical composition in FPs and UFPs would be precious for analysis of organic compounds in FPs and UFPs. Interestingly, organic and elemental carbon fractions were divided and discussed with meteorological and other gas pollutant data for source identification of FPs and UFPs.

Chapter 4 Characteristics of atmospheric black carbon (elemental carbon) in ultrafine and fine particles in urban environment, Japan

The characteristics of carbonaceous components especially have been focused on black carbon (char and soot). The carbonaceous components are crucial for adverse health effect and climate change, its characteristics are discussed with the different behavior between char and soot in ultrafine and fine particles for the better source apportionment.

Chapter 5 Evaluation of multichannel annular denuders for a newly developed ultrafine particle sampling system

For better understanding for the characteristics of UFPs, it is very important to measure not only particles but also gas phase target pollutants since some volatile chemical species exist in gas phase or particle phase due to gas-to-particle conversion (equilibrium). Hence, the collection method for gas phase pollutants using multichannel annular denuders was examined to apply this technique to INF sampler. Denuder coating and cleaning procedure is introduced in detail and the extraction efficiency, gas collection efficiency, and particle loss of UFPs within the denuders for ionic particles were studied and introduced in this Section.

Chapter 7 Summary and Concluding remarks

Importance of UFPs and possible sampling artifacts, UFPs measurement with gas and particle phase simultaneously and important knowledge having carbonaceous components and organic carbon species are discussed with possible

source identification of fine and ultrafine particles.

以上に要約したように、本論文は、最近健康影響の懸念から大きく話題となっている大気中超微小粒子の測定方法の開発とその応用、ならびに気候変動との関係で重要視されているブラックカーボン等の炭素系粒子の組成と挙動に関する研究であり、大気中における炭素粒子の発生源の推定への応用も行っており、学術的価値のみならず、工学的にも高く評価できる成果を挙げている。また、これに関わる内容の論文１編は既に学術誌に公表され、もう１編が受理され公表の段階に、さらに２編が審査中であり、本論文は博士（工学）の学位を授与するにふさわしい内容を備えていると判断し、当学位論文審査委員会は合格と判定した。