（ 続紙 1 ）

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論文題目	Study on the temporal and spatial variations of total OH reactivity and ozone production sensitivity in Tsukuba, Yokohama, and Kyoto in Japan		

（論文内容の要旨）

This research focused on three intensive field campaigns in contrasting urban sites in Japan. Total OH reactivity and trace species were detected concurrently to diagnose the air quality. Unknown trace species were quantified from the interpretation of total OH reactivity, and their influence on ozone production potential and ozone production regime was estimated. For the first time, heterogeneous effects on ozone production potential ware assessed quantitatively and NO (NO_X) concentration was found to be the key factor. Aerosol uptake effects also indicated a shift on the regime of ozone production, pointing to the significance of heterogeneous effects when considering ozone production mechanism and for mitigation policy. This dissertation contains 4 chapters as follows.

Introduction

The first half of Chapter 1 provided an overview of the background related to radicals, which is crucial to convert primary pollutants into secondary pollutants in the air. The whole contents focus on the generation, propagation, and termination of three types of radicals (OH, HO_2, and RO_2) in HO_X cycle. Second half of Chapter 1 introduced the set of instrumentation app- lied in field campaigns and specified current situation of field studies in terms of OH reactivity measurement.

Field measurements

Chapter 2 explained the details of three field measurements conducted in Tsukuba, Yokohama, and Kyoto. Each section firstly grasped the characteristics of measurement site, and then described in-situ observation from the perspective of trace species concentrations and total OH reactivity. Unknown trace species was described in the form of missing OH reactivity. Furthermore, information on the source of missing OH reactivity was characterized at each site. The last section in Chapter 2 synthesized all campaigns' results.

Ozone production sensitivity

Chapter 3 explored ozone production analysis based on the results from three field studies. As the main by-products of HO_X cycle, ozone's relationship with its precursors (VOCs and NO_X) and HO_X radicals was reported to be non-linear and complex. Mitigation of main precursors in the air cannot bring same reduction of ozone. Therefore, this chapter examined the mechanism of ozone production potential in different air conditions, quantified unknown trace species' contribution to oxidants generation, and further investigated the change of ozone production from heterogeneous effects, i.e. ozone reduction caused by the radicals loss via aerosol uptake.

The second part of Chapter 3 explained the results of ozone regime in the consideration of missing OH reactivity and aerosol uptake effects. VOC-limited regime was converted into NO_X-limited regime after ta- king the missing OH reactivity into account. Moreover, heterogeneous effects indicated identical transitional shift on the regime with missing OH reactivity. When making

stringent policies to control ozone, the consideration of influence from missing OH reactivity and heterogeneous effects would be necessary.

Conclusions

Chapter 4 summarized the major findings obtained from this study, proposed problems need to be considered in future for field studies, gave suggestions to solve air pollution problems, and achieve ozone mitigation effectively.

（論文審査の結果の要旨）

我が国の光化学オキシダント(Ox)レベルはその前駆物質の濃度が減少しているにもかかわらず明確な増加トレンドを示しており、その原因解明が急がれている。前駆物質としては揮発性有機化合物(VOC)が重要となるがその重要な発生源としては人為起源および植物起源が考えられている。人為起源由来のVOCは明確に削減が進んでいることから、植物起源のVOCの相対的な重要性が増加していると考えられている。また、Oxと同様重要な大気汚染物質であるPM2.5についてはOxと半相関的なトレンドを有することから、オゾン生成過程にエアロゾルとラジカルの相互作用が重要であるとの仮定を設定し、エアロゾルによる取り込み仮定を考慮したオゾン生成機構について考慮する必要性が出てきた。本論文では、過去に行った３つの集中観測（つくば、横浜、京都）結果を中心としてOH反応性、反応性微量成分計測、エアロゾル濃度、HO2のエアロゾルへの取り込み速度を計測し、その場における任意時刻の最大オゾン生成速度を計算し、その値のNOxおよびVOC濃度の感度を算出し、Ox生成に対するレジーム判定を行なっている。OHの未知反応性とエアロゾル取り込みがレジーム判定に与える影響について定量的に評価することで、精密なオゾン生成量の見積もりを可能としている。オゾン生成機構にエアロゾルとの相互作用を含めた本論文は初めての試みであり、その科学的な重要性はもとより、オキシダントの戦略的な制御を構築するためには極めて重要な知見を提供すると考えられられることから、地球環境学の発展に大きく貢献した。

よって本論文は博士（地球環境学）の学位論文として価値あるものと認める。また、令和３年８月16日、論文内容とそれに関連した事項について試問を行った結果、合格と認めた。

要旨公開可能日：　　年　　月　　日以降