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The Fundamental Requirements in the Application of Relaxed Eddy Accumulation Method for Measuring the Trace Gas Fluxes

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Abstract

It is well perceived that micrometeorological approach is one of the most reliable method for the quantification of vertical fluxes of trace components in the atmosphere. In this study, the feasibility of relaxed eddy accumulation (REA) method is discussed with respect to its reliability in the field application. Knowing that the use of micrometeorological approaches requires validation of analytical uncertainties involved, the problems and issues associated with its application are discussed to stimulate the proper employment of such technique in the field study.

Key words : Relaxed eddy, Micrometeorological, Flux, Atmosphere

1. INTRODUCTION

The environmental mobilization of trace constituents can be characterized with an aid of flux measurement techniques like eddy covariance (EC) method. Application of such method has, however, been limited to a few chemical species whose analysis can be made instantaneously by the employment of fast response sensor (i.e., > 1 Hz: CO_2, CH_4, NH_3 and so forth) (e.g., Businger and Oncley., 1990). Consequently, those methodological approaches had to be modified by and large for most chemicals whose analysis is not allowed by fast sensors. For instance, modified Bowen ratio (MBR) method has been employed successfully for the flux quantification of trace components like total gaseous mercury (TGM) (Kim *et al.*, 1995; Lindberg *et al.*, 1995).

This type of approach allowed to quantify TGM flux values by simultaneously measuring its concentration differences between different heights (above soil surface) and relevant micrometeorological parameters for the estimation of turbulent transfer coefficients (e.g., latent heat or momentum). By contrast, relaxed eddy accumulation (REA) method can also be considered as an excellent alternative technique which depends primarily on dividing vertically up- and downward moving air masses. The REA method can hence be used to provide flux values for a given component by measuring concentration differences in air masses of opposite vertical directions and relevant micrometeorological components (e.g., vertical wind speed). Although an attempt has lately been made for the flux measurements of exceptionally low traceable components (like reactive

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gaseous Hg: Lindberg *et al.*, 2002), this technique has been used extensively for natural volatile organic compounds (NVOC) including isoprene and/or terpene (e.g., Zhu *et al.*, 1999).

As such application requires highly precise and accurate quantification of concentration differences between up-and downward direction, its reliability can be questioned with respect to the analytical uncertainties. In this study, some limitations involved its application is discussed.

2. DISCUSSION

The reliability of those modified micrometeorological methods can be validated by accurately assessing the bias level of concentration differences (either between selected heights or between up-and downdraft air masses). To comply with such demand, the previous studies of the Hg flux measurements which typically relied on MBR method put much efforts to quantitatively explain the statistical significance of concentration differences between different heights. By conducting up to six replicate analyses of Hg concentration for each height level, Kim and Lindberg (1994) were able to accurately validate the significance of concentration differences between two heights above soil surface. Similarly to such approach, Kim *et al.* (2002, 2001) evaluated the uncertainty in measurements of concentration gradients in terms of percent gradient. On the basis of those previous studies, they reported that in most cases about 95% of concentration gradients quantified between two heights were statistically significant. The acquisition of highly reliable data sets may be feasible in the case of TGM, for its analysis can be made with excellent precision ($\sim$1%) and accuracy ($\sim$3%) (e.g., Kim *et al.*, 2002).

The REA method has been performed under various environmental settings but was not able to gain enough confidence from many respects; this in fact contrasts sharply with the case of the MBR method used previously for TGM flux measurements. For example, Gallagher *et al.* (2000) simultaneously conducted EC and REA methods for the measurements of CO_2 flux and verified an excellent agreement between the two methods. Despite the findings of such potentials for the REA method, its applicability on different target analytes needs to be tested further; this may be important because many of previous REA studies focused on NVOC flux measurements of which analysis can suffer due to relatively large uncertainties for micrometeorological approaches. In fact, the studies of Christensen *et al.* (2000) in which the basic aspects of NVOC measurement methods were evaluated for the derivation of their fluxes presented some referable information in such respect. These authors reported that NVOC can be measured with accuracy of about 10% and precision of about 2$\sim$6%. They hence suggested that concentration differences between the two different air masses can be assessed within the uncertainty range (precision) of 10 to 15%. Nonetheless, they did not provide information that can be used to estimate the proportion of statistically significant cases from measured concentration differences between the two different air masses.

Although the introduction of the REA method permitted to extend the applicability of micrometeorological methods, many of previous REA studies were not successful in providing quantitative information concerning the methodological reliability, especially with respect to the statistical significance in observed concentration differences between up-and downdraft air masses. In addition, as Edwards *et al.* (2002) addressed the point, contamination in either or both sampling lines to collect air samples for up-and downward direction may render this technique useless. Special care needs to be taken to prevent or reduce sources of such error (such as employment of clean Teflon sampling lines and confirmation of error sources). Considering all the complexities involved in the application of REA approach, there remain many uncertainties with respect to the validation of previously reported flux values of NVOCs. Therefore, future application of this method should firmly be based on the validation of its applicability, especially in the fundamental aspects of chemical quantification and statistical assessment.

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