Synthesized measurements of reactive nitrogen fluxes onto a forest using gradient and relaxed eddy accumulation method

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Introduction

In order to assess effects of reactive nitrogen on ecosystems, it is very important to estimate the rates of atmospheric deposition accurately. According to an intercomparison among inferential models to estimate dry depositions of reactive nitrogen, differences between the models reach a factor 2–3 and are often greater than differences between monitoring sites (Flechard et al., 2011). Therefore estimations of reactive nitrogen dry deposition by the models still have large uncertainties.

To improve the models, further understanding of dry deposition mechanisms by measurement-based studies is required. We introduce a synthesized measurement system for the fluxes of reactive nitrogen and relevant species in aerosol and gas phase onto a forest in FM Tama, western Tokyo.
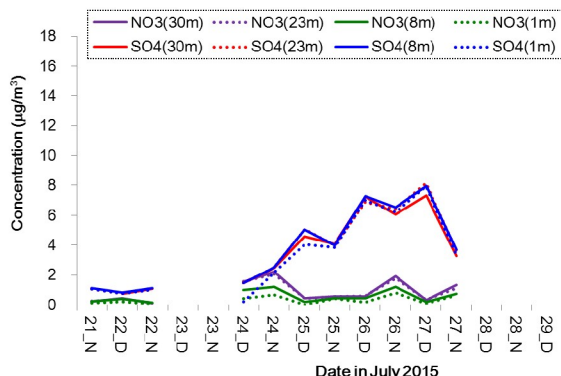
Synthesized measurement system by gradient and relaxed eddy accumulation (REA) method
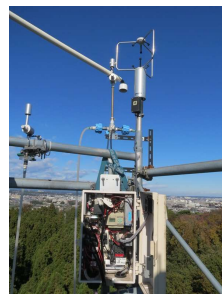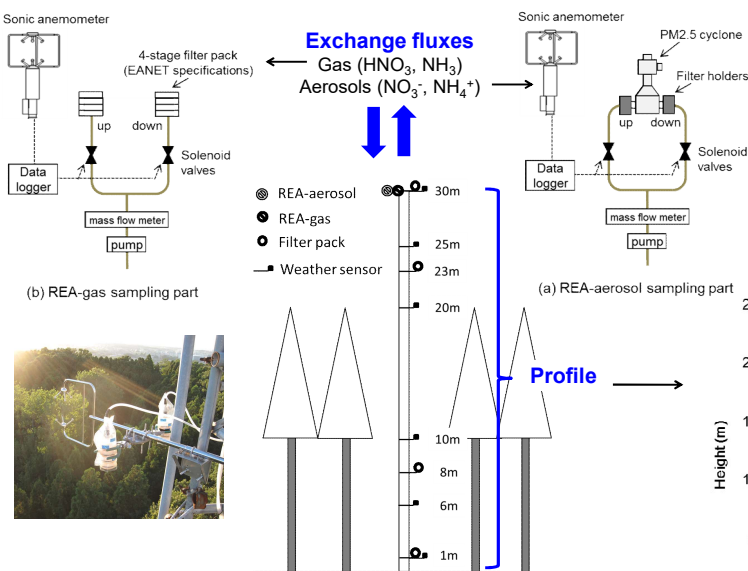
Experimental tower in FM Tama (western suburb of Tokyo, Japan)
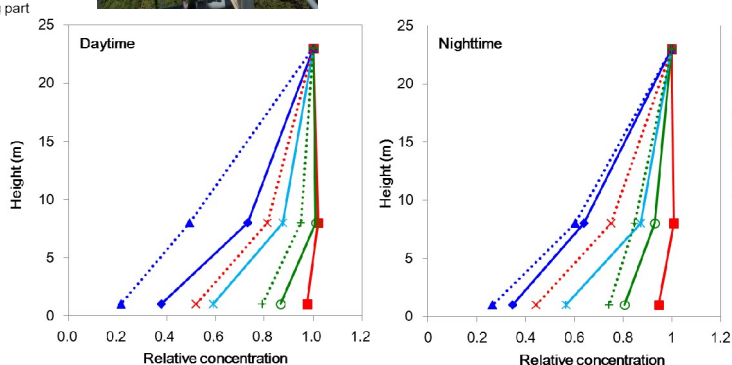
Mean vertical profiles of relative concentrations of $PM_{2.5}$ components (NO_3^-: NO3(F), SO_4^{2-}: SO4(F), NH_4^+: NH4(F)), NO_3^- in coarse aerosols (NO3(C)), HNO_3, SO_2 and NH_3. (Fig. 3)

The synthesized measurement system consists of vertical profile, aerosol flux (REA-aerosol) and gas flux (REA-gas) sampling parts.

➢ Vertical profile measurements

Filter packs at 1, 8, 23, 30 m ~ 2 heights above canopy, 2 heights below canopy
Concentrations of fine and course aerosol components, HNO_3, NH_3 and SO_2.

➢ Flux measurements

REA-aerosol: fluxes of $PM_{2.5}$ components, mainly NO_3^- and SO_4^{2-}
REA-gas: fluxes of HNO_3, NH_3 and SO_2
based on conditional sampling of updraft and downdraft of REA (relaxed accumulation) method

Experiments: the summer in 2015 (from 21 to 31 in July)

➢ During the period, the forest canopy was leafy and the leaf area index was estimated about 4–5.

➢ The samplings were continuously performed in the daytime (6:00–18:00) and in the nighttime (18:00–6:00) during the experiment, except rainy days.

Variations of concentrations of NO_3^- (NO3) and SO_4^{2-} (SO4) in $PM_{2.5}$ at 30m, 23m, 8m and 1m. "D" and "N" mean daytime sample (6:00-18:00) and nighttime sample (18:00-6:00), respectively. (Fig. 4)

Results and discussion

Deposition velocities (V_d) determined by REA measurements

	SO_4^{2-} in $PM_{2.5}$	NO_3^- in $PM_{2.5}$	HNO_3
Average	0.0	2.8	2.0*
29 night	0.0	1.4	1.3
30 night	-0.4	1.6	2.8

Unit: cm/s, V_d = Flux / Concentration, *: average in the night time

Theoretical Expectations

HNO_3 > SO_2 > NO_3^- in coarse > NO_3^- in $PM_{2.5}$ = SO_4^{2-} in $PM_{2.5}$

Results measured by the synthesized measurement system

HNO_3 > NO_3^- in $PM_{2.5}$ > SO_2 > NO_3^- in coarse > SO_4^{2-} in $PM_{2.5}$

➢ From the analysis of ion balance in $PM_{2.5}$ inorganic components, most of NO_3^- and SO_4^{2-} existed as NH_4NO_3 and $(NH_4)_2SO_4$, respectively. The discrepancy between theoretical expectations and the results is probably due to the differences of chemical properties between NH_4NO_3 and $(NH_4)_2SO_4$.

➢ Wyers and Duyzer (1997) indicates there is an effect of shifts in equilibrium between aerosol phase (NH_4NO_3) and gas phase (HNO_3, NH_3) near surfaces.

➢ Since the V_d of NO_3^- in $PM_{2.5}$ were close to that of HNO_3 measured by REA-gas, it is possible that NH_4NO_3 was quickly removed by the forest as well as HNO_3, due to the effect of shifts in equilibrium near surfaces.
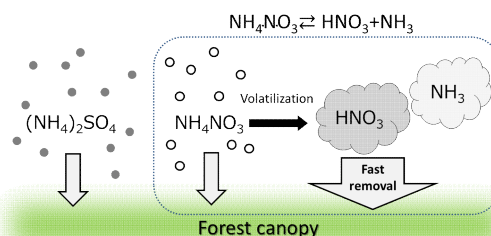
Conclusion

➢ Synthesized measurements of the fluxes of reactive nitrogen and relevant species in aerosol and gas phase using gradient and REA method were effective to better understand their deposition mechanisms.

➢ The measurements indicated that the effect of shifts in equilibrium between aerosol phase (NH_4NO_3) and gas phase (HNO_3, NH_3) near surfaces possibly enhanced dry deposition of NH_4NO_3.

➢ Current dry deposition ratios of NO_3^- in $PM_{2.5}$ is possibly underestimated, because the effect of shifts in equilibrium is usually not included in inferential models.

$NH_4NO_3 \rightleftarrows HNO_3 + NH_3$

Figure number and reference can be referred to the proceedings (4-page paper).