

Fluxes of NH_3 and HNO_3 over heathland in the Netherlands: implications for chemical interactions

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Abstract—The surface exchange of NH_3 and HNO_3 gases over heathland was measured with the aerodynamic gradient method in Leende, the Netherlands during April 25 and May 9, 1991. Due to reactions in the system NH_3 , HNO_3 and NH_4NO_3 , the NH_3 deposition flux is overestimated, while HNO_3 shows apparent emission fluxes mostly during the field experiment. Results reveal that NH_3 gradients are influenced slightly by HNO_3 and the HNO_3 gradients are strongly controlled by the NH_3 . A clear relationship between NH_3 and HNO_3 gradient is suggested by the Leende experiment. Before the late afternoon, NH_3 deposition gradients will cause HNO_3 upward gradients, meanwhile the NH_3 gradients themselves will become smaller. After the HNO_3 upward gradient reaches maximum, the NH_3 deposition gradients will be enlarged or the emission gradients depressed, while the HNO_3 upward gradients become smaller.

Keywords: dry deposition; ammonia; nitric acid; equilibrium, fluxes.

1 Introduction

Fluxes of NH_3 and HNO_3 have been measured separately or sometimes simultaneously in the past years (Duyzer, 1987; 1994; Wyers, 1992; Sutton, 1993; Erisman, 1993). For NH_3 , a bi-directional flux is characterized over farmlands (Sutton, 1993), while a perfect sink or a bi-directional flux with a weak emission component is found over semi-natural ecosystems (Wyers, 1992; Duyzer, 1987; 1994; Erisman, 1993). A major concern at present is the need to quantify the size of ammonia deposition to sensitive semi-natural ecosystems (Sutton, 1993). For HNO_3 , some representative results were obtained assuming chemical stationarity (Huebert, 1988; Harrison, 1989; Mueller, 1994).

Since there is possible atmospheric equilibrium between NH_4NO_3 aerosol and its gaseous precursors NH_3 and HNO_3 , chemical interference may influence the measurements of fluxes of these gases, especially for HNO_3 . Some evidences are available to show that such chemical interferences do exist (Huebert, 1988; Sutton, 1993). Huebert *et al.* (Huebert, 1988) found a steeper aerosol nitrate gradient than that of its vapor and an apparent emission of HNO_3 vapor sometimes. In Essex, England, an indication for possible interference was assumed when dry deposition velocity exceeded the maximum deposition velocity permitted by

turbulence (Harrison, 1989; Sutton, 1993). Significant negative R_c 's were observed when the data in Manndorf, South Germany (Mueller, 1993) was reprocessed. Actually, different effects of chemical interference were shown by Huebert *et al.* (Huebert, 1988), Harrison *et al.* (Harrison, 1989), and Müller *et al.* (Müller, 1993). In the study of Huebert *et al.* (Huebert, 1988), HNO_3 dry deposition was depressed and sometimes the direction of the HNO_3 flux was changed to apparent emission. In the studies by Harrison, Rapsomanikis *et al.* (Rapsomanikis, 1989) and Mueller *et al.* (Mueller, 1993), the HNO_3 fluxes are apparently enlarged.

The possible interactions among NH_3 and HNO_3 and the influence on their fluxes were modelled by Brost *et al.* (Brost, 1988) and Kramm *et al.* (Kramm, 1994). Since neither the structure of these chemical reactions nor the reaction rate constants are well known, sensitivity tests were made by using different chemical reaction constants. Brost *et al.* (Brost, 1988) showed that if the chemical lifetime of the dissociation equilibrium is 100s or less, there were appreciable effects upon the vertical gradients of the reactant species. For a slower chemical reaction, the species behave essentially conservatively and normal gradient-flux relationships apply.

In Leende, the Netherlands, a joint experiment on surface exchange of trace gases was conducted in 1991 (Duyzer, 1992). One of the objectives of the experiment was to determine dry deposition fluxes of all relevant nitrogen species and identify mechanisms regulating these fluxes. Some results on NH_3 fluxes in Leende have been reported (Wyers, 1992; Erisman, 1993). However, it is not clear if chemical interference violated the constant flux assumption. The purpose of this paper is to discuss the possible chemical interferences to NH_3 and HNO_3 fluxes, taking into account the equilibrium relationship between NH_4NO_3 aerosol and its gaseous precursors.

2 Experiment

From April 25 and May 10, 1991, a EUROTRAC/BIATEX joint field experiment was carried out over heathland in a nature reserve near Leende, the Netherlands. Details of this experiment were described by Duyzer *et al.* (Duyzer, 1992). The meteorological condition was given extensively by Duyzer *et al.* (Duyzer, 1992). HNO_3 and NH_3 concentrations were measured by two wet rotating denudes positioned at 0.93m and 3.47m with a time resolution of 40 minutes. The wet denuder has a precision of 5% (Keuken, 1988). Wind speed, temperature, friction velocity u^* and sensible heat flux were measured by a sonic anemometer (Duyzer, 1991, personal communication).

The theoretical background of the aerodynamic gradient method is extensively described by Fowler and Duyzer (Fowler, 1989) and Erisman (Erisman, 1990). The flux can be obtained from:

$$F = -u_* C_* , \quad (1)$$

where C_* is the eddy concentration, which is approximated by

$$C_* = \frac{ku_* [C(z_2) - C(z_1)]}{\ln[z_2 - d)/(z_1 - d)] - \varphi_h [(z_2 - d)/L] + \varphi_h [(z_1 - d)/L]} \quad (2)$$

Where d is the zero displacement height and φ_h is the stability correction for heat. The dry deposition velocity $Vd(z)$ can then be estimated from (Garland, 1977):

$$Vd(z) = -F/C(z) \quad (3)$$

The canopy resistance is usually deducted from the difference of the reciprocal of $Vd(z)$ and the sum of aerodynamic resistance Ra and the quasi-laminar resistance Rb .

$$Rc = 1/Vd(z) - Ra - Rb \quad (4)$$

As Rc tends to zero, $Vd(z)$ will approach the maximum deposition velocity Vd_{\max} . Physically, Vd_{\max} means that material can not be deposited faster at the surface than that permitted by turbulence. Therefore, Rc should be larger than or equal to zero and $Vd(z)$ is smaller than or equal to Vd_{\max} .

3 Results

The experiment can be clearly divided into three periods based on the concentration and flux magnitudes, as shown in Fig. 1. Drizzle and rain showers frequently occurred during April 30 and May 4, when very low concentrations and fluxes were observed until May 6. In general, opposite diurnal variations of NH_3 and HNO_3 fluxes are observed in Leende. The NH_3 fluxes are directed mostly towards the surface with maximum deposition fluxes occurring in the early morning. Occasionally emission of NH_3 is observed in the late afternoon under very dry conditions (Wyers, 1993). This bi-directional flux, though very weak, is considered as a behavior more typical for fertilized crop vegetation than for semi-natural vegetation. This heathland receives a high nitrogen load from atmospheric deposition. Upward

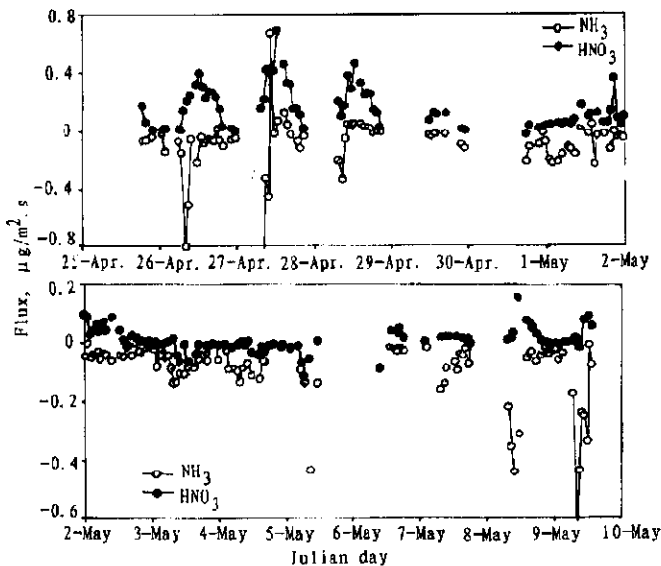


Fig. 1 NH_3 and HNO_3 fluxes in Leende, the Netherlands (April 25 – May 10, 1991)

HNO_3 gradients are found most of time during this experiment with maxima upward in the late afternoon. Downward gradients are observed only in a short period from May 3 to May 5, 1991, in which period the surface is wetted by drizzle and rain showers. It is believed that there is no direct emission of HNO_3 in the nearby environment, but the NH_3 gradient and the HNO_3 gradient are opposite in direction most of the time. Thus the upward gradients of HNO_3 are suspected to be a result of the reversible reaction between HNO_3 and NH_3 in the atmosphere.

The canopy resistances R_c for NH_3 exhibit diurnal variations with low resistances at night and high resistances at daytime (Fig. 2). Although R_c variations seem to be controlled by cuticular deposition at night and stomatal behavior at daytime, the magnitude of R_c is much lower than the prediction of cuticular and stomatal resistances, with much larger negative values at night. The canopy resistance for NH_3 is strongly related to the relative concentration gradient $\Delta C/C(z)$, as shown in Fig. 3. As the relative gradient of NH_3 increases, R_c changes from maximum positive to maximum negative consistently. In general, small gradient is suffered from serious measurement error. In Leende, many negative canopy resistances R_c 's, which mostly correspond to larger $\Delta C/C(z)$, are not caused by measurement errors but by other factors, probably by chemical reaction. While the larger positive R_c 's, corresponding smaller $\Delta C/C(z)$, are suffering from larger measurement errors.

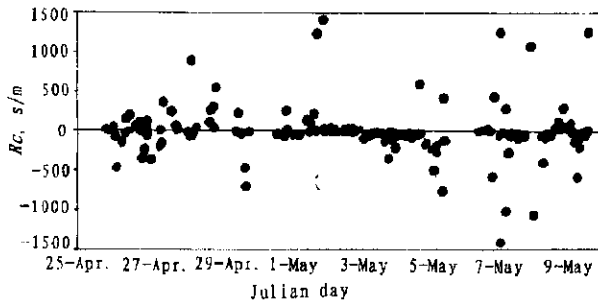


Fig. 2 Canopy resistance of NH_3 in Leende, the Netherlands, April 25—May 9, 1991

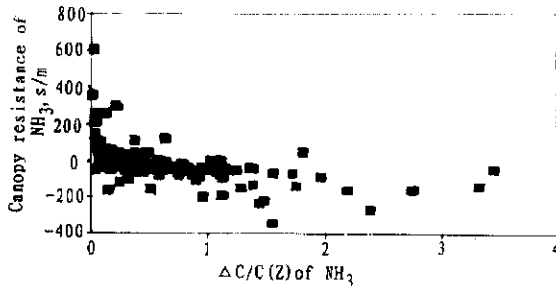


Fig. 3 Canopy resistances of NH_3 as a function relative concentration gradient of NH_3

The humidity dependences of NH_3 and HNO_3 flux are seen in Fig. 4. NH_3 generally has flux directed to the surface, the higher the relative humidity RH, the larger the deposition flux of NH_3 will be. Small emission flux of NH_3 is observed at RH less than 60% (Wyers, 1993). Contrast to NH_3 flux, HNO_3 generally has fluxes directed away from surface, the lower the relative humidity, the larger the upward flux will be. Most deposition fluxes of HNO_3 are observed at RH larger than 60% during the experiment in Leende.

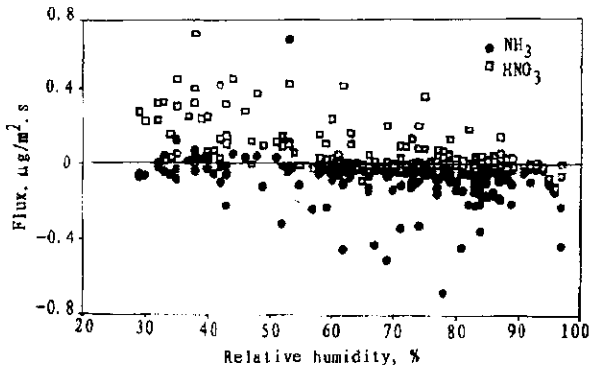


Fig. 4 NH_3 and HNO_3 fluxes as a function of relative humidity in Leende, the Netherlands (April 25–May 10, 1991)

After a data selection procedure to satisfy the constant flux requirement, the average NH_3 flux and deposition velocity are estimated as $-75 \text{ ng}/(\text{m}^2 \cdot \text{s})$ and 2.6 cm/s for all data including both deposition and emission fluxes, or $-80 \text{ ng}/(\text{m}^2 \cdot \text{s})$ and 2.8 cm/s for deposition fluxes only. The estimates for flux and velocity are slight higher than those obtained earlier for heathland (Duyzer, 1993) and for forest (Wyers, 1994). The mathematical average R_c for NH_3 is 21 s/m . While for the average obtained by $1/(\text{Avg}(R_c^{-1}))$, inverse of average R_c^{-1} , it is -39 s/m . For HNO_3 , it is impossible to give an estimate for its flux and deposition velocity due to apparently unrealistic concentration gradients.

4 Discussion

The upward fluxes of HNO_3 and the negative canopy resistances R_c for NH_3 imply that some factors have seriously enlarged the NH_3 concentration gradient and depressed the HNO_3 gradient, or even changed its direction. The possible chemical influence on the HNO_3 gradient by NH_3 was suggested already in some field experiments by Huebert, Lake, Delany and Brost (Huebert, 1988) and Sutton, Pitcairn and Fowler (Sutton, 1993). Brost, Delany *et al.* (Brost, 1988) and Kramm and Dlugi (Kramm, 1994) showed by numerical simulation that NH_3 , HNO_3 and NH_4NO_3 aerosol can influence each other, but the extent is strongly dependent on the chemical equilibrium time scale compared with the turbulent time scale. If the chemical equilibrium in air is reached instantaneously, the fluxes of HNO_3 and NH_3 will not be influenced by this equilibrium (Kramm, 1994).

4.1 Equilibrium relationships between NH_4NO_3 aerosol and its gaseous precursors

Although equilibrium between NH_4NO_3 aerosol and its gaseous precursors is believed to exist in atmosphere (Stelson, 1982; Harrison, 1994; Allen, 1989), the occurrence of the equilibrium is challenged by some serious departures at high humidity and low temperature (Allen, 1989; Pio, 1993). The relationship among NH_3 , HNO_3 and NH_4NO_3 in Leende is shown in Fig. 5, in which the ratio of the measured gaseous HNO_3 and NH_3 concentration product K_m to the theoretical prediction K_e is presented. Any ratio K_m/K_e not equal to unity means departure from equilibrium. Strong diurnal variations; much larger than unity in night and slightly lower in the late afternoon, can be seen in Fig. 5, indicating absence of equilibrium in Leende. The diurnal variation of K_m/K_e is more or less the same as that of NH_3 concentrations, implying that NH_3 influences the equilibrium greatly. In general, the NH_3 , HNO_3 and NH_4NO_3 aerosol reaction system tends to aerosol condensation in Leende; conditions allowing aerosol evaporation only occur for short time in the late afternoon.

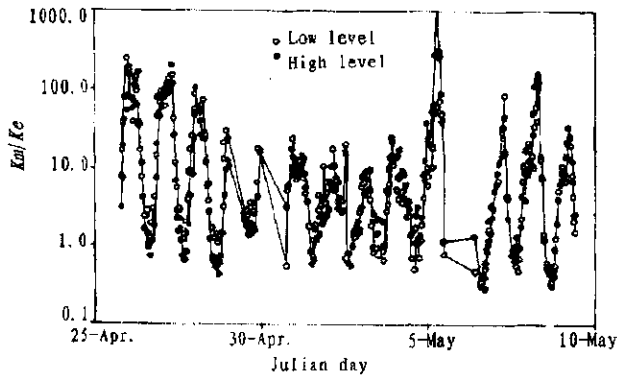


Fig. 5 The variation of K_m/K_e in Leende, the Netherlands (April 25–May 9, 1991)

Regression analysis for soluble aerosol components reveals that NH_4^+ and NO_3^- show a very good linear relation. The molar ratios $\text{NH}_4^+/\text{NO}_3^-$ vary between 0.72 and 1.89 with an average of 1.09, indicating that all NO_3^- exists as NH_4NO_3 aerosol. Assuming that all sulfate is presented as ammonium sulfate $(\text{NH}_4)_2\text{SO}_4$ or $(\text{NH}_4)\text{HSO}_4$ and that the remaining NH_4^+ combines with NO_3^- , there is no enough NH_4^+ to accommodate all NO_3^- to form NH_4NO_3 . However, a good linear relation between remaining NH_4^+ and NO_3^- is found:

$$([\text{NH}_4^+] - 2 \cdot [\text{SO}_4^{2-}]) = -0.00847 + 0.6856[\text{NO}_3^-], \quad r = 0.947 \quad (5)$$

$$([\text{NH}_4^+] - [\text{SO}_4^{2-}]) = -0.00612 + 0.8938[\text{NO}_3^-], \quad r = 0.939 \quad (6)$$

The total or remaining NH_4^+ is correlated poorly with Cl^- . The gaseous HNO_3 molar concentration is much higher than HCl , implying that the reaction $\text{NaCl} + \text{HNO}_3 = \text{NaNO}_3 + \text{HCl}$ is not fast enough and that HCl is not in equilibrium with NH_4Cl aerosol. Thus, both aerosol and gaseous concentrations indicate that the reversible reaction system among ammonium salts and their gaseous precursors is shifted to form ammonium salts. This apparently irreversible reaction system offers a possibility to explain HNO_3 and NH_3 flux behaviors.

4.2 The possible influence of irreversible reactions between HNO_3 and NH_3 on their exchange fluxes

In Leende, clear relations between the fluxes are observed in two periods of the experiment; April 26–29 and May 7–8, which are shown in Fig. 1. The flux of NH_3 shows same the variation as that observed in other experiments; maximum deposition fluxes in the night or early morning, occasional emission in midday. The flux of HNO_3 shows emission flux mostly in the experiment. Basically, NH_3 concentration is much larger than HNO_3 and its huge gradient can not be easily changed in direction by HNO_3 . Due to low concentration in atmosphere, gradient of HNO_3 will be influenced by the NH_3 gradient if the reaction between NH_3 and HNO_3 is fast but not fast enough to reach equilibrium on a time scale of turbulence. A possible explanation for the HNO_3 flux variation can be given according to the irreversible chemical reaction mathematically described in equations 7 and 8. As the equations, the HNO_3 flux will change in the way opposite to NH_3 flux variation. In the early morning, the NH_3 flux gradually increases to its maximum deposition. Meanwhile, the strong NH_3 deposition gradient will gradually depress the HNO_3 deposition gradient. In Leende, it causes the HNO_3 gradient to change direction to apparent emission. Later, when the NH_3 deposition flux is getting smaller, the HNO_3 emission flux still increases. HNO_3 upward gradients will reach maxima when $(k_1[\text{NH}_3][\text{HNO}_3])_H - (k_1p\text{NH}_3)[\text{HNO}_3])_L$ is equal to zero (subscript H and L means high level and low level measurement height). This phenomenon is usually observed in the afternoon when the NH_3 deposition flux is lowest or when NH_3 is emitted. After that, the HNO_3 upward gradient will decrease and the NH_3 deposition flux will remain relatively constant or increase slightly because of atmospheric stability. As equation 8 indicates, the NH_3 gradient will be influenced by the HNO_3 gradient too. The influence is probably only significant in the afternoon. At that time, HNO_3 has its strongest upward gradient, while NH_3 has either a small deposition flux or an emission flux. Therefore, the NH_3 emission gradient will be depressed or the deposition gradient enlarged because of an upward HNO_3 gradient. It seems that NH_3 gradient can result in a large change in the HNO_3 gradient. However, it is not clear whether the upward gradient of HNO_3 in the late afternoon can result in the large negative R_c 's for NH_3 in the morning.

$$\Delta\text{HNO}_3(t) = \Delta\text{HNO}_3(t_0) - \int \{ (k_1[\text{NH}_3][\text{HNO}_3])_H - (k_1[\text{NH}_3][\text{HNO}_3])_L \} dt, \quad (7)$$

$$\Delta\text{NH}_3(t) = \Delta\text{NH}_3(t_0) - \int \{ (k_1[\text{NH}_3][\text{HNO}_3])_H - (k_1[\text{NH}_3][\text{HNO}_3])_L \} dt. \quad (8)$$

Fig. 6 and Fig. 7 show the relationship between NH_3 and HNO_3 fluxes statistically as the functions of their gradient ratio. The relation of flux and gradient ratio can be divided into four parts (for example; $F(\text{HNO}_3) - \Delta\text{NH}_3/\Delta\text{HNO}_3$) in Fig. 6:

- (1) upper left: HNO_3 emission, NH_3 deposition,
- (2) lower left: HNO_3 deposition, NH_3 emission,
- (3) upper right: HNO_3 emission, NH_3 emission,
- (4) lower right, HNO_3 deposition, NH_3 deposition.

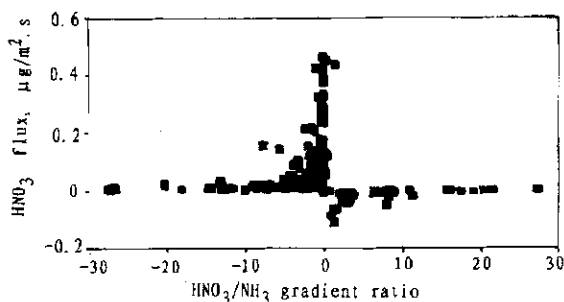


Fig. 6 HNO₃ flux as a function of NH₃/HNO₃ gradient ratio, April 25—May 9, 1991, in Leende, the Netherlands

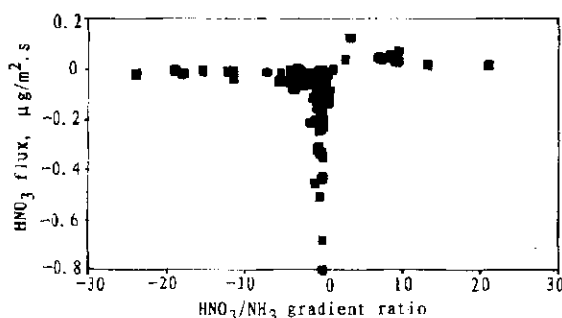


Fig. 7 NH₃ flux as a function of HNO₃/NH₃ gradient ratio, April 25—May 9, 1991, in Leende, the Netherlands

HNO₃ fluxes are mostly located in the upper left part, which is believed to be caused by NH₃ deposition. Some are located in the lower right part, which suggests co-deposition of HNO₃ and NH₃. HNO₃ has deposition fluxes most during May 3—5 with frequent rain showers occurring. In this period, the relationship among HNO₃ and NH₃ is not clear. Apparent co-emission of NH₃ and HNO₃ is clearly seen in Fig. 7, showing the NH₃ flux as a function of $\Delta\text{HNO}_3/\Delta\text{NH}_3$. As Fig. 1 shows, the apparent co-emission for NH₃ and HNO₃ mostly occurs in the late afternoon when the NH₃ gradient is small and the HNO₃ upward gradient is large.

In principle, the diagram showing flux as a function of concentration gradient ratio can give some qualitative information about gradient enlargement or depression. For example, in Fig. 6, 7, in the upper left part of the figure, the NH₃ deposition gradient is depressed greatly because the HNO₃ flux has changed to apparent emission. In the upper right part, both HNO₃ and NH₃ emission fluxes are depressed. In the lower left part, HNO₃ deposition flux is enlarged, while the NH₃ emission flux is depressed. In lower the right part, both HNO₃ and NH₃ deposition fluxes are depressed.

Fig. 8, showing HNO₃ flux as a function of $(\text{NH}_3/\text{HNO}_3)_H/(\text{NH}_3/\text{HNO}_3)_L$ ratio, is also designed to describe the relationship between NH₃ and HNO₃. The ratio of NH₃/HNO₃ is a

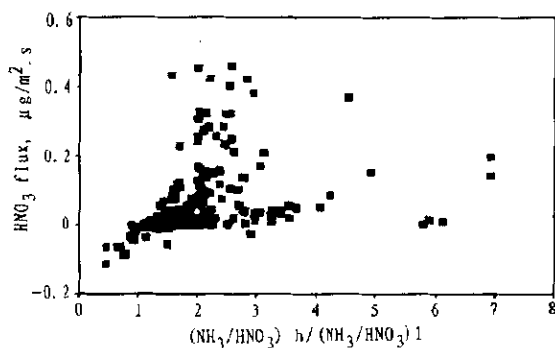


Fig. 8 HNO_3 flux as a function of concentration ratio of NH_3 to HNO_3 , the ratio is defined as $(\text{NH}_3/\text{HNO}_3)_H / (\text{NH}_3/\text{HNO}_3)_L$

measurement of atmospheric acidity and the HNO_3 gradient is found to be related with the vertical distribution of the ratio. The ratio R is defined to be equal to $(\text{NH}_3/\text{HNO}_3)_H / (\text{NH}_3/\text{HNO}_3)_L$. Three situations are found in Fig. 8:

$R < 1$ or $(\text{NH}_3/\text{HNO}_3)_H < (\text{NH}_3/\text{HNO}_3)_L$. In this case, NH_3 is usually emitted and HNO_3 is deposited because air acidity is higher at higher measurement level.

$R > 2$ or $(\text{NH}_3/\text{HNO}_3)_H \gg (\text{NH}_3/\text{HNO}_3)_L$. The strong downward gradient of NH_3 results in an upward gradient of HNO_3 .

When R is in the range between 1 and 2, the direction of HNO_3 gradient is uncertain. The HNO_3 gradient can be directed downward or upward dependent on NH_3 gradient. In Leende, HNO_3 is mostly deposited when $R < 1.5$ and emitted when $R > 1.5$.

5 Conclusion

The variations in NH_3 fluxes over heathland are directed mostly to the surface with occasional emission in the late afternoon under very dry conditions with RH less than 60%. This variation was observed previously by Erisman and Wyers (Erisman, 1993) and Wyers, Vermeulen, and Slanina (Wyers, 1992). Many negative Rc 's for NH_3 , i. e. $Vd > Vd_{\text{max}}$, are found at high humidity. The average flux and dry deposition velocity Vd for NH_3 are $-75 \text{ ng}/(\text{m}^2 \cdot \text{s})$ and 2.6 cm/s with an average canopy resistance Rc of -39 s/m . The significant negative canopy resistance implies that average flux and deposition velocity are overestimated. HNO_3 fluxes are mostly directed away from the surface, indicating serious chemical interference.

NH_3 and HNO_3 are not in equilibrium with NH_4NO_3 aerosol at all with much more gases present in the gas phase than theoretically predicted. This phenomenon suggests that NH_3 and HNO_3 flux will be influenced by their atmospheric reactions.

Taking into account the irreversible reaction between NH_3 and HNO_3 , the results emphasize the existence of chemical interference on fluxes of NH_3 and HNO_3 in Leende. The

NH_3 flux is influenced slightly by HNO_3 and the HNO_3 flux is strongly controlled by the NH_3 flux. A clear relationship between NH_3 and HNO_3 fluxes is suggested by this experiment. Before the late afternoon, NH_3 deposition gradient will cause an upward HNO_3 gradient. Meanwhile the NH_3 gradient will become smaller. After the HNO_3 upward gradient reaches maximum, the NH_3 deposition gradient will be enlarged or an emission gradient depressed, after which the HNO_3 gradient becomes smaller. This relationship is partly supported by the observed variation of canopy resistances for NH_3 .

Acknowledgements— The author gratefully acknowledge Dr. G. P. Wyers, ECN the Netherlands for providing the data set in Leende in the Netherlands. The project was supported by Trans-Centure Training Programme Foundation for Talents by the State Education Commission.

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