Comment on " Diurnal fluxes of HONO above a crop rotation " by Laufs et al

This paper presented results of flux measurements of HONO over an agriculture field using the gradient method. Based on the averaged diurnal profile, the authors calculated the HONO flux which was then used in correlation studies to explore the contributing mechanism. Photosensitized heterogeneous conversion of NO2 on soil surfaces were suggested as the major contributor to the HONO flux based on the correlation results and a local parameterization of HONO flux was also proposed. Overall, this is a well-designed study trying to answer the challenging question about atmospheric HONO source. However, there are some important issues that need to be addressed before it is accepted for publication in ACP.

Major issues:

(1) HONO flux calculation

According to Eq (2), the calculated flux depends on the gradient of HONO at two heights. If the difference between such values is too small, e.g. comparable to the systematic difference of instruments ~2-13% as shown in Fig. 2, it may invalidate most discussions. I would like to see the difference in both absolute and relative term (ΔHONO and ΔHONO/HONO)

$$F_{z_{ref}} = -\kappa \cdot u_* \cdot \frac{\partial c(HONO)}{\partial[\ln(z-d) - \Psi_{(z-d)/L}]}$$

The authors used averaged values to interpret the flux data: "*To interpret the flux data for each measurement campaign, a diurnal average was calculated by the formation of one-hour means from the whole measurement period.*" My question is how did you average $\Psi_{(z-d)/L}$ and $\Psi_{(z-d)/L}$, etc, since their expression is different between stable and unstable conditions. In principle, it is a question whether we should first calculate the flux at each time and then do the averaging, or first do the averaging of individual parameters and then calculate the flux. Can the authors address this issue and try to make calculation for both cases?

My last question is if the effect of chemistry can be neglected in the calculation of HONO fluxes. We can make a simple HONO budget expression around noon time as follows,

$\partial HONO/\partial t = \partial F/\partial z + S$

in which the change of HONO concentration ($\partial HONO/\partial t$) is subject to the gradient of flux ($\partial F/\partial z$) and the photolytic loss term (S). If $\partial HONO/\partial t << S$, then the contribution of S should be comparable to flux ($\partial F/\partial z$) and cannot be neglected.

(2) Soil surface emission

The diurnal course of soil temperature strongly depends on the depth. Here the authors used soil temperature at 5 cm in their calculations. I would suggest using soil surface temperature as in Su et al. (2011) which is more relevant for soil-atmosphere exchange. Figure 1 of Su et al. also suggests that HONO produced by photo-sensitize reaction (on the surface) is subject

to the temperature dependent equilibrium. Since the peak of soil surface temperature appears earlier than that of deeper soil (see the following figure, Jury and Horton 2004), the correlation with HONO fluxes might be improved.

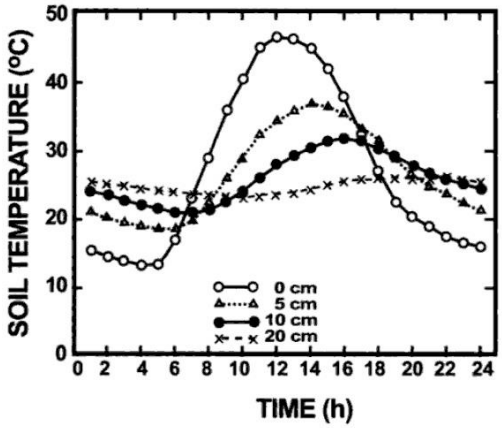

Figure 5.17 Diurnal variations in temperature measured at different depths in a loam soil. (After Yakuwa, 1945.)

Minor comments:

Ln 22: "unusually high" suggests that the measured values is higher than the expected values. Many of the references, however, don't really have an expected value from modeling or budget analysis. Thus I suggest modifying the text or limiting the references to those with budget analysis. The following references should be included into the reference list (Su et al 2008, Li et al. 2012, Yang et al. 2014).

Ln 30: "bacterial production of nitrite in soil", it is better to say "biogenic production of nitrite in soil"

Ln 42: " calculated daytime HONO sources, determined from HONO levels exceeding theoretical photostationary state (PSS) values, showed high correlations with the photolysis rate coefficient *J(NO2)* or the irradiance and NO2 concentration (Elshorbany et al., 2009; Sörgel et al. 2011b; Villena et al., 2011; Wong et al., 2012; Lee et al., 2016)." So far as I know, Su et al. (2008) is the first study performing such correlation analysis and is unfortunately missing from the reference list.

Reference:

W. A. Jury, R. Horton, *Soil Physics*. (Wiley, ed. 6th, 2004).

Li, X., Brauers, T., Häseler, R., Bohn, B., Fuchs, H., Hofzumahaus, A., Holland, F., Lou, S., Lu, K. D., Rohrer, F., Hu, M., Zeng, L. M., Zhang, Y. H., Garland, R. M., Su, H., Nowak, A., Wiedensohler, A., Takegawa, N., Shao, M., and Wahner, A.: Exploring the atmospheric chemistry of nitrous acid (HONO) at a rural site in Southern China, Atmos. Chem. Phys., 12, 1497-1513, 10.5194/acp-12-1497-2012, 2012.

Su, H., Cheng, Y. F., Shao, M., Gao, D. F., Yu, Z. Y., Zeng, L. M., Slanina, J., Zhang, Y. H., and Wiedensohler, A.: Nitrous acid (HONO) and its daytime sources at a rural site during the 2004 PRIDE-PRD experiment in China, JGR, 113, 10.1029/2007jd009060, 2008.

Yang, Q., Su, H., Li, X., Cheng, Y., Lu, K., Cheng, P., Gu, J., Guo, S., Hu, M., Zeng, L., Zhu, T., and Zhang, Y.: Daytime HONO formation in the suburban area of the megacity Beijing, China, Science China Chemistry, 57, 1032-1042, 10.1007/s11426-013-5044-0, 2014.