Interactive comment on "Study of the unknown HONO daytime source at an European suburban site during the MEGAPOLI summer and winter field campaigns" by V. Michoud et al.

General comments:

In this manuscript HONO budget in the suburban site of Pairs was studied and possible HONO sources were thoughtfully discussed. The parameters including OH and J value measured in this campaign are valuable and relatively comprehensive for HONO budget analysis. In the analysis, major known sources were included and several unknown sources were reasonably proposed by correlation analysis. Thus, I think this manuscript as a generally high quality research and recommend publication in ACP despite of some concerns listed in the specific comments.

Specific comments:

(1) Mathematic dilemma

The result of this study is mainly supported by the correlation between $S_{unknown}$ and $J(NO_2)$. However, $S_{unknown}$ is calculated by subtracting the known HONO source from all HONO sinks which is dominated by $(HONO)*J_{HONO}$, see equation 4 and figure S1.1 and S1.2. Considering the high correlation between J(HONO) and $J(NO_2)$, positive correlation between $S_{unknown}$ and $J(NO_2)$ is expected mathematically. Although it is commonly accepted in the previous publications that the positive correlation between $S_{unknown}$ and $J(NO_2)$ indicates a photolytic characteristic of the unknown HONO sources. I recommend that we think the positive correlation a little more conservatively without a precursor (NO_2/NO_3^-) clearly proposed regarding this mathematic dilemma.

(2) Transport and mixing

The wind speed from west and southwest is more than 4 m/s, see Fig 1 and Fig 3. The transport and vertical mixing of HONO could not be neglected. Even under the assumption of well mixed air mass in both clean marine region and Pairs city, wind induced vertical mixing alone could be a significant HONO sink in such high wind speed, not mention of the radiative induced mixing. Lack of vertical mixing

in the PPS and $S_{unknown}$ calculation can result in large uncertainty in correlation analysis. This should be considered in this manuscript at least.

(3) $S_{unknown}$ could be different in different air mass

I recommend separate the days of different wind directions considering the differences in wind speed and chemical environment. As stated above, wind induced mixing could be essential as a HONO sink when west and southwest wind apply. While wind induced mixing will be smaller in case of north wind. Besides, the HONO chemistry in clean marine air mass represents an aged or a background condition while the HONO chemistry in urban air mass is similar to that of a direct emission or fresh aged mixed air mass. For instance, fresh sea salt aerosol from west may act as a sink of HONO while secondary particle from north is considered as a HONO source. By the way, the role of the total surface area of aerosol was not discussed in the result.

(4) Uncertainty in PPS_2 and second thought on $S_{unknown}$ calculation, see equation (1) and equation (2)

Besides vertical mixing, some parameters in this study could further contribute to the uncertainty of PPS_2 and $S_{unknown}$ calculation. V_{HONO}, V_{NO2}, Va, HONO/NOx ratio are highly variable. The choice of them is easily challenged. For instance, 0.008 are the upper limit of HONO/NOx ratio reported in the literature. It is inappropriate even for the fresh urban air mass which takes almost two hours (comparing to HONO lifetime of around 20 min.) to reach the research site according to a wind speed of 2 m/s. In addition, the assumption of no vertical gradient for HONO concentration is unrealistic and will also increase the uncertainty of PPS_2 calculation. Again, in the early morning and later afternoon, the $P_{emission}$ is comparable to the $P_{unknown}$ (see Figure S1.1 and S1.2), which means that the uncertainty of $P_{emission}$ is transferrable to $P_{unknown}$. At last, due to the small J(HONO) in the early morning and later afternoon, the PPS assumption is more invalid comparing to the noon time when lifetime of HONO is around 15 min, $P_{unknown}$ is then suffering from higher uncertainty. In all, I recommend using PPS_1 instead of PPS_2 to calculate $S_{unknown}$ and also restrain $S_{unknown}$ in the noon time in correlation analysis.

(5) Difference in correlation coefficients and slopes, see Fig14 and Table 3
Under the assumption of photolytic characteristic of the unknown source, high correlation between $S_{unknown}$ and J (NO_2) should not fail. However, low correlation efficient is found in the days when a pretty good agreement is found between PPS

and measured HONO (see line 5-8 in23660). This indicates again that uncertainty in $S_{unknown}$ calculation affect correlation analysis in a bad way. Thus, both correlation coefficients and slopes need a double check with a renew $S_{unknown}$ recommended above. Even though, the slopes are similar in some extent in picking days (says R^2 bigger than 0.8) considering the uncertainty in calculation. The slope around 240 in the winter and 80 in the summer, however, could be a clue for HONO precursor.

(6) Writing corrections

Detailed concerns are not listed. However, technical corrections are needed after further thought on the uncertainty of $S_{unknown}$ calculation and correlation analysis.