## **Response to Reviewer #CC1, Ezra Wood**

### **Comment**

Under what range of humidity values were the experiments that quantified how well the denuder removes gas-phase compounds (sections 2.1.1 and 3.1.1) conducted? Friedrich et al. (AMT, 13, 5739–5761, 2020, https://doi.org/10.5194/amt-13-5739-2020) demonstrated degraded performance of a similar activated carbon denuder to various nitrogen oxides under humid conditions compared to dry conditions. Ideally there is no humidity dependence for the denuder at hand to the range of organic compounds studied, but it would be reassuring for this to be experimentally determined.

### **Response (our first response):**

Many thanks for this comment. We did not consider this humidity effect before. Our gas denuder experiments were conducted under dry conditions, and we observed near unity removal efficiency for the tested VOC, similar to those reported for nitrogen oxides with dry flows (Friedrich et al., 2020). But as reported by Friedrich et al. (2020), the performance of the gas denuder may decrease as the humidity increases, and in their experiments, the removal efficiency decreased to ~65% for some NOy species. Furthermore, a "used" denuder was found to release NOx (converted from stored NOz at the surface) in humid air (Friedrich et al. 2020). Testing for such effects also for organics would be very important for our study, and for other studies using a similar gas denuder as we used, e.g. in the chemical analysis of aerosol online (CHARON) inlet (Eichler et al., 2015) and the extractive electrospray ionization time-of-flight mass spectrometer (EESI-TOF) system (Lopez-Hilfiker et al., 2019). In addition, there are studies that reported similar degraded adsorption capability of activated carbon denuders to VOC in humid conditions, and this effect depends on the coating and design of the denuders (Li et al., 2020; Li et al., 2021).

In practice, dryers are typically used for ambient aerosol measurements, and thus potential humidity effects can be minimized even if the denuders show such behavior. But it is indeed very interesting and important to investigate the humidity-dependent removal efficiency of organic compounds for the denuder used in our study. We expect to be able to conduct such tests during the next months when we have access to the necessary instrumentation and will thus be able to give a more detailed answer still during this review process.

References:

Eichler, P., Müller, M., D'Anna, B., and Wisthaler, A.: A novel inlet system for online chemical analysis of semi-volatile submicron particulate matter, Atmos. Meas. Tech., 8, 1353-1360, 10.5194/amt-8-1353-2015, 2015.

Friedrich, N., Tadic, I., Schuladen, J., Brooks, J., Darbyshire, E., Drewnick, F., Fischer, H., Lelieveld, J., and Crowley, J. N.: Measurement of NOx and NOy with a thermal dissociation cavity ring-down spectrometer (TD-CRDS): instrument characterisation and first deployment, Atmos. Meas. Tech., 13, 5739-5761, 10.5194/amt-13-5739-2020, 2020.

Li, X., Zhang, L., Yang, Z., He, Z., Wang, P., Yan, Y., and Ran, J.: Hydrophobic modified activated carbon using PDMS for the adsorption of VOCs in humid condition, Separation and Purification Technology, 239, 116517, https://doi.org/10.1016/j.seppur.2020.116517, 2020.

Li, Z., Jin, Y., Chen, T., Tang, F., Cai, J., and Ma, J.: Trimethylchlorosilane modified activated carbon for the adsorption of VOCs at high humidity, Separation and Purification Technology, 272, 118659, https://doi.org/10.1016/j.seppur.2021.118659, 2021.

Lopez-Hilfiker, F. D., Pospisilova, V., Huang, W., Kalberer, M., Mohr, C., Stefenelli, G., Thornton, J. A., Baltensperger, U., Prevot, A. S. H., and Slowik, J. G.: An extractive electrospray ionization time-of-flight mass spectrometer (EESI-TOF) for online measurement of atmospheric aerosol particles, Atmos. Meas. Tech., 12, 4867-4886, 10.5194/amt-12-4867-2019, 2019.

**Response (our current response):**

During the past months, we had a chance to conduct some further experiments to assess the performance of the gas denuders under humid conditions. A Vocus proton transfer reaction (PTR), equipped with a long time-of-flight mass spectrometer, and the same VOC cylinder as described in section 2.1.1 were used during the experiments. We tested the performance of two different gas denuders under dry (<1%) and humid (73$\pm$2 %) conditions, as the best and worst scenarios, respectively, with a working flow rate of 1 L $min^{-1}$. Note that 13 different VOC species with a concentration of ~10 ppb for each specie (thus ~130 ppb in total, which is much higher than ambient conditions) was used during the experiment. But, we only plotted alpha-pinene as an example, because they all showed comparable variations during the experiment.

The results of this experiment are shown in Figure S5 and summarized in Table S4, which have been added to the revised manuscript. In general, better removal efficiency was obtained under dry compared to humid conditions for both the "new" and "old" gas denuders (i.e. lower concentrations of alpha-pinene during the "VOC + denuder" stages in light red area vs. in light blue area). In addition, during the continuous exposure experiment (Figure S5b), we observed faster performance degradation of the gas denuder under humid compared to dry conditions, indicating extra caution is needed during long-term usage of these gas denuders.

Overall, as the reviewer pointed out that the humidity effect is very important, we added more discussions in Section 3.1.1 line 274, to inform the readers about the results of our recent experiment.

*"**Table S4.** Description and performance of the two tested gas denuders. The time series are shown in Figure S5.*

| *Gas denuder* | *#1* | *#2* |
|---|---|---|
| *Description* | | |
| *Usage count* | *> 20* | *1* |
| *Regeneration count* | *> 20* | *1* |
| *Status before test* | *regenerated* | *regenerated* |
| *Usage time* | *~5 month* | *~1 week* |
| *Description* | *"old"* | *"new"* |
| *Performance (removal efficiency in terms of ~10 ppb alpha-pinene)* | | |
| *Dry* | *87%* | *94 %* |
| *Humid* | *76 %* | *84 %* |
| *continuous (~18-20 h) exposure to VOC under* | | |
| *Dry* | */* | *92% to 87 %* |
| *Humid* | *92% to 81%* | */* |

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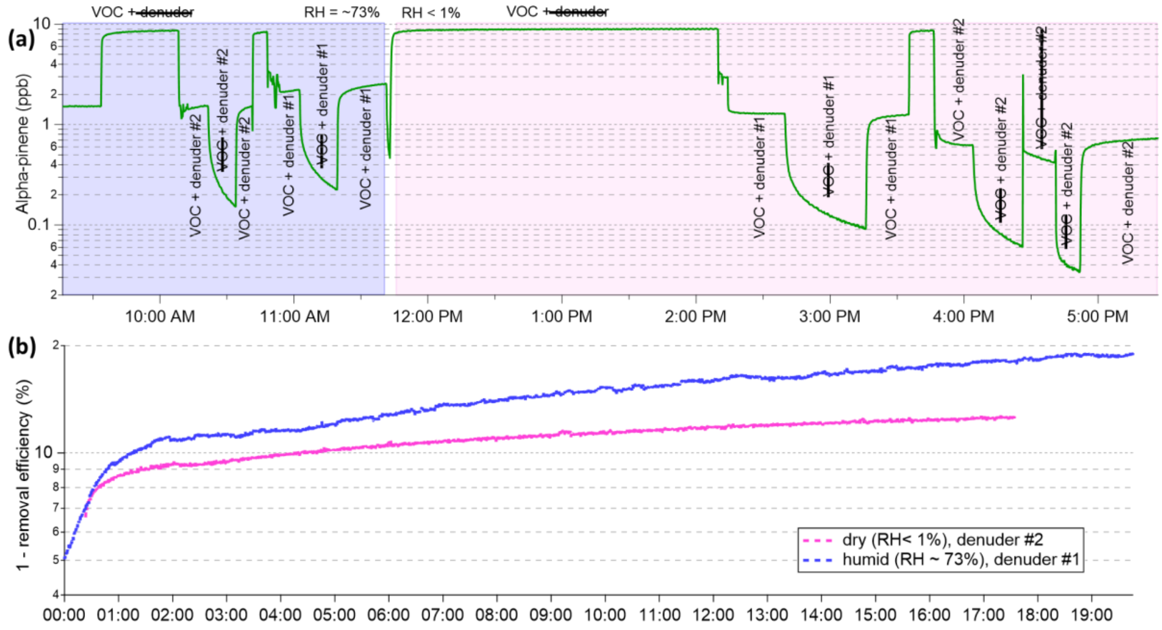

***Figure S5.*** *Time series of alpha-pinene during (a) gas denuders test under dry (light blue area) and humid conditions (light red area) and (b) continuous exposure to VOC under dry and humid conditions. Because the rest VOC standards showed comparable variation during this experiment, alpha-pinene is plotted here as an example to show the humidity effects on the performance of gas denuders. In panel (a), the text explains the condition of each stage, i.e. different combinations of VOC flow and gas denuder, and strikethrough means without. The performance of these two gas denuders is summarized in Table S4."*