Authors' Response to Referee Comments

We appreciate the overall positive response of the Referee and we would like to thank for his constructive comments and helpful suggestions on the manuscript, which helped us to further improve the clarity of the paper. Below, we give detailed responses (in blue) where appropriate.

Dave Nelson (Referee)

This is an excellent paper describing a truly impressive accomplishment: the dramatic miniaturization of a laser based trace gas monitor with very little sacrifice to measurement accuracy. The results are convincing and the paper is very well written and should certainly be published. I have a few small suggestions for the authors to consider but the suggestions are not mandatory.

1. Taking advantage of the surprisingly narrow water line is very clever. However, the discussion describing why this line is narrow is not clear. Does it simply have a small broadening coefficient or is it narrow because of Dicke narrowing which does not, I think, involve energy level spacings. Or is it both? It would be nice to clarify this discussion or remove it if the explanation is not clear.

We are thankful for pointing out this weakness of the discussion. Indeed, the description was misleading and therefore we revised it to avoid any confusion. Just to clarify: The linewidth of an absorption at given pressure is determined by two different contributions, the Dicke narrowing and the pressure broadening. Pressure broadening is due to collisions, which perturb internal motion. Changes in rotational energy, especially for linear molecules, contribute sizably to linewidth. In general, broadening is small for high-*J* transitions, because these states have energy separations of the order of 300 cm^{-1} from other rotational states to which they can make collisional transitions. It is therefore difficult to absorb this energy in translational motion (kT ~200 cm^{-1}). (Eng *et al.*, Appl. Phys. Lett. (1972), 21, 303). On the other hand, a significant narrowing can be observed if (i) the quantum state of the molecule is not affected by collisions and (ii) the mean free path is smaller than the wavelength of the proving radiation. The high *J*-states have long rotational lifetime and thus exhibit also a significant collisional (Dicke) narrowing. This, however, manifests in the Doppler widths of spectral lines at higher pressures, which can be narrower than the width expected from the Maxwell-Boltzmann distribution (Giesen *et al.*, J. Mol. Spec. (1992), 153, 406-418). The collisionally narrowed line profiles, however, cannot be fitted to the Voigt function with the Doppler width expected from the Maxwell-Boltzmann distribution. The treatments of such line profiles requires more sophisticated profile functions that include the effect of soft- and hard-collision (e.g. Ngo *et al.*, Phil. Trans. R. Soc. A (2012), 370, 2495–2508). Since this effect is mainly dominating at gas pressures < 200 hPa, we decided to not mention its contribution in our paper, which is primarily focused at atmospheric pressure. Therefore, we modified the text in our manuscript correspondingly:

"This can be explained by inefficient collisional relaxation due to the wide energy separation (~300 cm^{-1}) for high-J states of H_2O (Eng et al., 1972; Giesen et al., 1992). Considering the typical energies of translation motion (kT ≈200 cm^{-1}), it is obviously difficult to take up the transition energies from such rotational states. Therefore, the perturbation of these high rotational energy states by collision remains low, which results in low pressure broadening, so that the H_2O absorption line at ambient pressure appears as narrow as it would be at 0.1 atm.

2. It is a little surprising to me that the detector temperature has a strong effect on the reported mixing ratio. Do the authors have an explanation for this? Changes in linearity or bandwidth, perhaps?

We share this surprise, but the observations strongly support this effect. Actually, we observed that mainly the shape of the signal profile was changing rather than the signal amplitude. We contacted the detector supplier and they pointed out that these small footprint preamp packages (SIP) have insufficient heatsinking capabilities, and it is mandatory to have additional, external heatsink to guarantee sufficient heat dissipation. We think that the Peltier-element used to cool the detector chip is overstrained, and that it loses temperature stabilization, resulting in the observed symptoms.

3. The statement near the end of Section 3.1 that the remaining temperature artifacts "most likely reflect the susceptibility of the entire electronics to abrupt temperature changes" seems unsupported. Do the authors have a reason for believing that the problem could not be due to an optical effect? If so that should be stated. If not, then perhaps it would be better not to speculate.

Yes, we have good reason. Since the optics is very compact, it is rather straightforward to induce temperature fluctuations affecting almost exclusively the optical elements only. Conversely, this is also true for the electronics. The "hot-spots" that we were able to identify are the potentiometers used to adjust the shape and amplitude of the current pulses for the detector. At less extent, a resistor element used in the reference current source can also be contributing to the instabilities/drifts of the laser driving current.

Although, these effects are minor, the broad linewidth of the methane absorption line is highly susceptible to any slight change in the laser emission. Currently, we are developing a newer version of our laser driver, where the potentiometers are replaced by digital signals.

4. Figure 6 shows a comparison between the compact QCL monitor and a Picarro monitor. The scale is so large that it is difficult to see the discrepancies. It would be useful to add a trace that shows the difference between the mixing ratios reported by the two instruments.

We added a plot to Fig.6 showing the difference between the mixing ratios reported by the two instruments.