

1 We thank Anonymous Referee 3 for their careful reading of our manuscript and appreciate their thoughtful  
2 feedback. We have edited the manuscript to incorporate their critiques, which we feel has improved the overall  
3 quality of this work.

4  
5 Below, Referee comments are presented in bold, and our responses are written in regular type. Line numbers in  
6 responses refer to the “track changes” version of the manuscript, which will be uploaded separately. For explicit  
7 clarity in text changes, added text will be in blue font, while deleted and/or moved text in ~~red-strikethrough~~ for  
8 clarity. Changes to figures will only be outlined in the responses to the Referee.

## 10 Anonymous Referee 3

11 **The manuscript describes a gas phase HCL sensor, based on tunable infrared laser direct absorption**  
12 **spectroscopy (TILDAS). The authors highlight the importance of HCL in the atmosphere, describing its**  
13 **influence. They describe the difficulties of monitoring HCL, it is “sticky” and results in such effects as**  
14 **long instrument response times. They describe current monitoring techniques, their strengths and**  
15 **weaknesses, and why their approach will be of benefit.**

16 **The authors first describe the TILDAS sensor design, followed by techniques to minimize sticky behavior**  
17 **of HCL on extraction, an “inertial inlet” and active passivation. They also describe procedures for**  
18 **validation, field testing and data analysis.**

19 **The authors present results for different configurations, such as with and without passivation and**  
20 **humidity effects, They present field data and compare sensitivity to other published set-ups. They discuss**  
21 **problems with HCL particulate and nitric acid.**

22 **They present 7-8 pptv at 1 Hz and  $3\sigma$  limit of detection ranging from 21-24 pptv. For longer**  
23 **averaging times, the highest precision obtained was 0.5 pptv and  $3\sigma$  limit of detection of 1.6 pptv at**  
24 **2.4 minutes. These values are competitive compared to other optical techniques, which are considered**  
25 **more complicated to set-up. I think the manuscript should be published.**

26 **I believe the manuscript requires minor revisions and clarifications.**

- 27 • **The title needs to be considered. I find it misleading. TILDAS is not a novel spectroscopic**  
28 **approach. It is the first application of TILDAS to HCL. Should be clarified to reader or**  
29 **manuscript changed.**

30 The title has been modified to “Using Tunable Infrared Laser Direct Absorption Spectroscopy for  
31 ambient hydrogen chloride detection: HCl-TILDAS”.

- 32 • **There is little detail on optical configuration of set-up. If it is new and custom made, more**  
33 **information can be given here.**

34 As pointed out by the Referee, the TILDAS technique is now many years old. Further detail is  
 35 extensively given to these topics (including TILDAS measurement principle and structural schematic  
 36 diagram) by McManus et al. (2011, 2015) for the TILDAS design used in this work:

37 McManus, J. B., Zahniser, M. S., and Nelson, D. D.: Dual quantum cascade laser trace gas instrument  
 38 with astigmatic Herriott cell at high pass number, *Appl. Opt.*, 50, A74,  
 39 <https://doi.org/10.1364/AO.50.000A74>, 2011.

40

41 McManus, J. B., Zahniser, M. S., Nelson, D. D., Shorter, J. H., Herndon, S. C., Jarvis, D., Agnese, M.,  
 42 McGovern, R., Yacovitch, T. I., and Roscioli, J. R.: Recent progress in laser-based trace gas instruments:  
 43 performance and noise analysis, *Appl. Phys. B*, 119, 203–218, [https://doi.org/10.1007/s00340-015-6033-](https://doi.org/10.1007/s00340-015-6033-0)  
 44 0, 2015.

45 These publications are now properly cited, and the reader is now more clearly directed these references  
 46 for additional detail (lines 137-140). Additionally, we have included more details on the HCl specific  
 47 attributes for the instrument used in this publication in Sect. 2.2.1 (lines 150-165).

48

- 49 • **There is little detail to spectral fitting. They do talk about background subtraction. But, error can**  
 50 **also come from the spectra fit. I think more detail should be given here.**

51 Spectral fits are non-linear least squares fits of a  $\sim 1 \text{ cm}^{-1}$  spectral window, using a nonlinear least-  
 52 squares fit that includes a polynomial baseline. Peak location is fixed using a frequency-locking  
 53 algorithm based upon the deep methane lines. Pressure and temperature are included in the fit to  
 54 account for pressure broadening and rovibrational state populations, respectively. These details have  
 55 been added to Sect. 2.2.1 (lines 150-165).

- 56 • **Re Methane measurement. Do you have a LOD or sensitivity for this measurement? it appears in**  
 57 **plot, methane can fluctuate by approx 2-3 ppb in a few seconds. is this real? If yes, why not see**  
 58 **these types of fluctuations with HCL**

59 The line strengths used for the methane measurement are approximately 10x less than for HCl, and is  
 60 therefore expected to be a less precise measurement. For the data presented in Fig. 6, standard  
 61 deviations for the high concentration average 3 ppbv, and 2 ppbv while sampling the lower  
 62 concentrations. This would result in an approximate LOD of 4-9 ppbv. However, it should also be  
 63 clarified that the methane data presented in Fig. 6 for the submitted manuscript used normalized  
 64 concentrations that represent a change in methane from a high concentration from a zero-air cylinder  
 65 ( $\sim 2250 \text{ ppbv}$ ) to a lower concentration as measured by ambient air ( $\sim 2220 \text{ ppbv}$ ); the normalization  
 66 was done for timescale of signal decay calculation comparisons. However, we have modified Fig. 6 to  
 67 restore the methane mixing ratios as originally observed, and to make it clear to the reader we are not

68 operating near these estimated limits of detection. Further characterization of this methane signal is  
69 outside the scope of this paper.

70 • **ISORROPIA II.. a few sentences on theory are not in referred section**

71 This has been corrected; the appropriate references now appear in the References section.

72 • **The conclusion seems more like an outlook, except for first sentence.**

73 More summary detail has been added to the Conclusion section (lines 555-570).

74 **Individual line comments/typos;**

75 **line 107... typo "it is has"**

76 This has been corrected (line 108).

77 **line 138... What is exact wavelength of laser and tuning range?**

78 Laser radiation probes the strong R(1) H<sup>35</sup>Cl line (2925.89645 cm<sup>-1</sup>) of the (1-0) rovibrational absorption band  
79 near 3.4 μm (lines 154-155). The spectral window (2925.80 to 2926.75 cm<sup>-1</sup>) has been added to Sect. 2.2.1  
80 (lines 150-153).

81 **line 160... "gas phase via acid displacement" can you add a reference for this. There are references**  
82 **earlier, but they don;t seem to fit this.**

83 We have added a reference to Roscioli et al., 2016 which demonstrates this effect for nitric acid. We have also  
84 clarified that we are referring to a mechanism analogous to that which occurs with aerosol (e.g, Beichert and  
85 Finlayson-Pitts, 1997), line 172.

86 Roscioli, J. R., Zahniser, M. S., Nelson, D. D., Herndon, S. C., and Kolb, C. E.: New Approaches to Measuring  
87 Sticky Molecules: Improvement of Instrumental Response Times Using Active Passivation, J. Phys. Chem. A,  
88 120, 1347–1357, <https://doi.org/10.1021/acs.jpca.5b04395>, 2016.

89 Beichert, P. and Finlayson-Pitts, B. J.: Knudsen Cell Studies of the Uptake of Gaseous HNO<sub>3</sub> and Other Oxides  
90 of Nitrogen on Solid NaCl: The Role of Surface-Adsorbed Water, J. Phys. Chem., 100, 15218–15228,  
91 <https://doi.org/10.1021/jp960925u>, 1996.

92

93 **line 401 "It is well established that HCl and particulate chloride (pCl- 401 ) exist together in dynamic**  
94 **equilibrium" can you add refernce here.**

95 We have inserted citations for the following references (lines 440-442):

96 Beichert, P. and Finlayson-Pitts, B. J.: Knudsen Cell Studies of the Uptake of Gaseous HNO<sub>3</sub> and Other Oxides  
97 of Nitrogen on Solid NaCl: The Role of Surface-Adsorbed Water, *J. Phys. Chem.*, 100, 15218–15228,  
98 <https://doi.org/10.1021/jp960925u>, 1996.  
99 Brimblecombe, P. and Clegg, S. L.: The solubility and behaviour of acid gases in the marine aerosol, *J.*  
100 *Atmospheric Chem.*, 7, 1–18, <https://doi.org/10.1007/BF00048251>, 1988.  
101 Clegg, S. L. and Brimblecombe, P.: The dissociation constant and henry's law constant of HCl in aqueous solution,  
102 *Atmospheric Environ.* 1967, 20, 2483–2485, [https://doi.org/10.1016/0004-6981\(86\)90079-X](https://doi.org/10.1016/0004-6981(86)90079-X), 1986.  
103 Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for  
104 K<sup>+</sup>-Ca<sup>2+</sup>-Mg<sup>2+</sup>-NH<sub>4</sub><sup>+</sup>-Na<sup>+</sup>-SO<sub>4</sub><sup>2-</sup>-NO<sub>3</sub><sup>-</sup>-Cl-H<sub>2</sub>O aerosols, *Atmospheric Chem. Phys.*, 7, 4639–4659,  
105 <https://doi.org/10.5194/acp-7-4639-2007>, 2007.

106 **line 484 How is nh3 measured here?**

107 A Los Gatos Research ammonia analyzer was used. This is now explicitly stated on line 536.

108 **Figure 10. Is the time on a) and b) the same. Not clear. y scale strange**

109 The timescale on a) and b) are the same. Vertical guidelines have been added to make this clear. The y-axis  
110 labels have been modified