

1 We thank Anonymous Referee 1 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.

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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~~
8 clarity. Changes to figures will only be outlined in the responses to the Referee.

10 Anonymous Referee 1

11 **Comments to the Author:**

12 **The paper describes a commercial TILDAS instrument for measuring hydrogen chloride in ambient air**
13 **and demonstrate the ability of sampling methodology to minimize inlet artefacts. Due to the “sticky”**
14 **behavior of HCl gas, quantitative sampling remains a challenge for current approaches. To improve**
15 **instrument response to changes in HCl gas concentration, a custom-fabricated quartz virtual impactor is**
16 **used to replace particle filters to avoid excess surface-mediated interactions with filters, and the heating**
17 **and PFBS coating methods are employed to improve transmission. Its performance validates that the**
18 **sampling method is effective for reducing HCl “sticky” behavior. Overall, the paper is well written, with**
19 **detailed characterization in the lab as well as reliable performance in the field sampling. I recommend this**
20 **paper for publication in AMT after the following minor revisions.**

21 **General comments:**

22 **Section 2.2.1: The technique description of the TILDAS device is not clear, and more technical details**
23 **need to be added, such as measurement principle, structural schematic diagram, etc.**

24 As pointed out by Anonymous Referees 2 and 3, the TILDAS technique is now many years old. Further detail
25 is extensively given to these topics (including TILDAS measurement principle and structural schematic
26 diagram) by McManus et al. (2011, 2015) for the TILDAS design used in this work:

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

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30 McManus, J. B., Zahniser, M. S., Nelson, D. D., Shorter, J. H., Herndon, S. C., Jarvis, D., Agnese, M., McGovern,
31 R., Yacovitch, T. I., and Roscioli, J. R.: Recent progress in laser-based trace gas instruments: performance and
32 noise analysis, Appl. Phys. B, 119, 203–218, <https://doi.org/10.1007/s00340-015-6033-0>, 2015.

33 These publications are now properly cited, and the reader is now more clearly directed to these references for
34 additional detail (lines 138-140 in “track changes” manuscript). Additionally, we have included more details on
35 the HCl specific attributes for the instrument used in this publication in Sect. 2.2.1 (lines 150-165).

36 **Section 2.2.2: The custom-fabricated quartz virtual impactor can effectively remove the large particles (>**
37 **300 nm diameter) in the sampling line, which was approximately 13% of the total volumetric flow. Only**
38 **gas molecules and small particles (< 300 nm diameter) can flow into the TILDAS instrument. Please**
39 **explain how does the impactor work and how is the ratio of flow rate obtained?**

40 The inertial inlet is interfaced with the instrument scroll pump, as seen in Fig. 1, which pulls air through the
41 inertial inlet via two paths: 1) through a waste flowpath that does not pass through the TILDAS, and 2) through
42 the TILDAS. Sample air that enters the inertial inlet is accelerated through a critical orifice into a low-pressure
43 region (< 100 torr). Once in the low-pressure region, particulate separation occurs as follows: large particles (>
44 300 nm diameter) have large forward momentum and maintain their forward flow into the waste flow path
45 (approximately 13% of the total volumetric flow, dictated by a separate critical orifice placed in the waste-flow
46 path). Meanwhile, gas molecules and particles with an approximate diameter < 300 nm have less inertia and
47 can make the 180° turn necessary to continue along the sample flow path through the TILDAS instrument
48 (approximately 87% of the total volumetric flow). The resulting flow rate through the instrument was
49 determined by the size of the critical orifice in the inertial inlet and cell pressure (set to approximately 40 torr).

50 We have revised this passage to clarify how particle separation occurs within the inertial inlet (lines 175 – 185).
51 We have also referred the reader to Fig. 1 to visualize sample flow paths (line 187). Additionally, we have
52 labeled the ambient pressure and low-pressure regions of the inertial inlet in Fig. 1.

53 **Section 3.1: The performance of HCl TILDAS is evaluated in the lab with dry zero air as well as in the**
54 **field with HCl-scrubbed sample air, and its precision and LOD are superior to the previously reported**
55 **methods. More technical details need to be added to explain how does the instrument achieve better**
56 **performance? Did the authors perform long-term measurements of a fixed concentration of HCl gas?**
57 **This approach can better represent its real performance.**

58 The better performance of the HCl TILDAS is achieved using a long pathlength (200 m), measuring absorptions
59 in the mid-infrared by probing the fundamental ro-vibrational absorption band (which have a much larger cross-
60 section than in the near-IR), and reducing light and dark noise levels to $<5 \times 10^{-6}$ equivalent absorbance in 1-
61 second. We now provide these details in lines 310-313.

62 Regarding long-term measurements of a fixed concentration of a fixed gas, we performed a series of permeation
63 source additions and removals across ~28 hours, resulting in 55, 10-min permeation source additions and
64 subsequent 20-min background measurement periods. We note the permeation source concentration over this
65 period was 4.1 ± 0.3 ppbv, and that permeation source concentration variability correlated closely with
66 laboratory air-conditioning. Nevertheless, the average standard deviation calculated for the last five minutes of
67 each permeation source additions was found to be 8 ± 2 pptv, while the average standard deviation of the last

68 five minutes of background periods was calculated as 7 ± 1 pptv, demonstrating nearly identical precisions
69 while sampling blanks or fixed HCl concentrations. These details have been added to Sect. 3.2.1, lines 356-360.

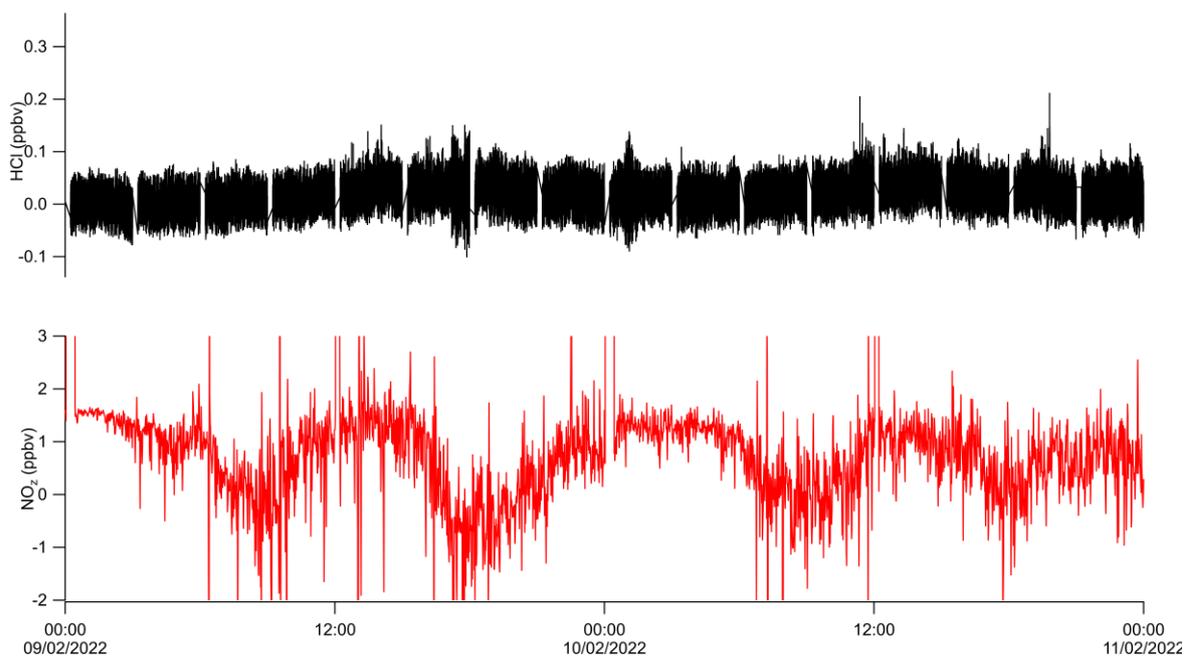
70 **Section 3.3.2: There is an obvious offset about 0.07 ppbv (shown in Figure 8) before addition of nitric acid**
71 **to the passivated sample inlet flow. Please explain the reason for the offset signal.**

72 The data presented were not blank subtracted. Figure 8 has been revised to use blank subtracted data.

73 **Section 3.4: The maximum concentration of HCl in field observation is about 0.1 ppbv shown in Figure**
74 **9(a). But the HNO₃ concentration of 4 ppbv may cause an increase of 0.08 ppbv of HCl. How to evaluate**
75 **the error of atmospheric HCl concentration caused by HNO₃? And the influence of a potential leak on the**
76 **measurement of HCl gas concentrations during observation needs to be clearly evaluated.**

77 The intention of the laboratory HNO₃ addition experiments (Sect. 3.3.2) was to demonstrate the potential of an
78 acid-displacement-induced interference, as ambient sampling will be further complicated by additional strong
79 acids, such as H₂SO₄, that may also cause rapid acid displacement reactions with HCl sorbed on inlet surfaces.
80 As discussed in Sect. 3.3.2, the magnitude of the resulting HCl plume will be a function of how much HCl is
81 taken up by instrument surfaces. This emphasizes the importance of reducing the amount of surface HCl
82 available for off-gassing, though this effect will likely be dampened for in situ sampling, in which ambient
83 strong acid concentrations change much more gradually. We have added a comment to emphasize the
84 importance of reducing HCl sorption (lines 491-492).

85 For the described field measurements (Sect. 3.4), the largest potential source of HCl for coating the inlet
86 surfaces will be the regular permeation source or standardized HCl cylinder additions for 10 minutes every 3
87 hours. Unfortunately, the absolute magnitude of sorbed HCl is difficult to quantify, as Fig. 7 demonstrates the
88 complex relationship between stickiness and humidity, although regular measurement of inlet response times
89 provides a metric by which it can be monitored, and the inlet subsequently cleaned if deemed an issue. As
90 discussed in Sect. 3.4, we do not see evidence of significant interference from HNO₃ (as estimated via NO_z)
91 from our field results, given the differences in the diurnal profiles between HCl and NO_z. Further, a preliminary
92 comparison of data from the winter OSCA campaign shows virtually flat HCl signals while NO_z maintains a
93 diurnal profile, suggesting our results are likely not affected to a detectable degree by, at least, HNO₃ (please
94 note that a 1 ppb offset has been added to the NO_z data, and that the NO_y, NO₂, and NO data that were used to
95 calculate NO_z have not been QC-checked at the time of this response):



96

97 We additionally do not rule out future measurements from being affected by this potential interference. The
 98 dependence on inlet condition, however, means that we are able to estimate any additional uncertainty on the
 99 reported HCl using the regularly measured inlet response Tau values. Further, we are currently testing the
 100 implementation of temperature ramping the inlet to near 100 °C following additions of HCl standards to remove
 101 potential surface HCl from the inlet caused by the HCl standard addition, although a full field assessment of the
 102 impact of these temperature ramps has not yet been performed. We have added text to the Conclusions section
 103 about the potential improvements that this could have (lines 563-565).

104 Concerning the influence of a potential leak of HNO₃, we do not employ HNO₃ permeation sources during
 105 ambient HCl sampling, and therefore are not susceptible to such a leak.

106 **Specific comments:**

107 **Page 4, L137: The references could not be found in this manuscript.**

108 This has now been corrected, and these references have been properly added to the Reference section.

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

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

115 **Page 14, L382-387: The influence of humidity on the measurement bias of HCl concentrations is only**
116 **reported at 60% RH. In fact, the relative humidity of atmosphere is often much higher than this value.**
117 **Therefore, the authors need to give the relationship between the measurement bias and the relative**
118 **humidity, so that the reader can clearly grasp it.**

119 The pertinent passage in this comment refers to the effects of HCl standards being injected into the inlet under
120 ambient relative humidities during the OSCA campaign. To that end, the mean and standard deviation
121 presented were calculated for relative humidities between 60-93%, and compared with the mean and standard
122 deviation of dry, compressed air at relative humidities below 20%. We have revised the sentence to clarify that
123 the statistics and data presented in Fig 7b include these humidity ranges, and that the values are not solely
124 obtained under an RH of 60% (lines 421-424).

125 **Page 18, L490: The data should be modified to 20 June 2021.**

126 The date has been modified (line 542).