

Laminar gas inlet – Part 2: Wind tunnel chemical transmission measurement and modelling

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Abstract. Aircraft-based measurements of gas-phase species and aerosols provide crucial knowledge about the composition and vertical structure of the atmosphere, enhancing the study of atmospheric physics and chemistry. Unlike aircraft-based aerosol particle sampling systems, the gas loss mechanisms and transmission efficiency of aircraft-based gas sampling systems are rarely discussed. In particular, the gas transmission of condensable vapors through these sampling systems requires

15 systematic study to clarify the key factors of gas loss and to predict and improve gas sampling efficiency quantitatively. An aircraft gas inlet for aircraft-based laminar sampling of condensable vapors is described in part 1 (Yang et al., 2024), which describes the inlet dimensions, flow analysis and modelling, along with initial gas transmission estimates. Here we test and characterize the complete inflight sampling system for gas-phase measurements of H_2SO_4 in a high-speed wind tunnel, and conduct detailed computer fluid dynamics (CFD) simulations to assess inlet performance under a range of flight conditions.

20 The gas transmission efficiency of H_2SO_4 through different sampling lines was measured using Chemical Ionization Mass Spectrometry (CIMS), and the experimental results are reproduced by the CFD simulations of flow and mass diffusion using a mass accommodation coefficient, $\alpha_i = 0.70 \pm 0.05$ for H_2SO_4 on inlet lines. The experimental data and simulation results show consistently that gas transmission efficiency increases with an increased sampling flow rate. At $Re \sim 2300$, the overall inlet transmission is $16 \pm 6\%$ for H_2SO_4 at ground and high altitude. The simulation results further indicate that sampling

25 efficiency can continue to improve to a certain level after the sampling flow enters the turbulent flow regime, up to Reynolds numbers, $Re \sim 6000$. A decrease in transmission is predicted only for higher Re numbers. These results challenge the widely held assumption that laminar flow core sampling is the best strategy for sampling condensable vapors. The gas-phase H_2SO_4 transmission efficiency can be optimized (increased by a factor ~ 2) by minimizing residence time, rather than maintaining laminar flow; this benefit extends to other condensable vapors and applies over the full range of operating conditions of the

30 aircraft inlet system. For a sticky species ($\alpha_i > 0.25$), the laminar diffusivity is important to predict the transmission efficiency via the aircraft inlet section, while for less sticky species ($\alpha_i < 0.25$) the gas-phase diffusivity plays a minor role in predicting the gas transmission efficiency in the sampling line.

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1 Introduction

Knowledge of the composition of the atmosphere and its change over time is relevant to public health, air quality and climate.

35 Atmospheric composition research requires measurements and understanding of both its gas-phase and aerosol constituents. A particular analytical challenge is the in-situ aircraft-based measurement of condensable vapors, which critically contribute to both aerosol formation through nucleation and its early growth processes. During sampling, condensable vapors such as sulfuric acid can be efficiently lost to pre-existing aerosol surfaces and the inner walls of inlet lines. Such losses alter and distort downstream measurements of the properties of sampled gas and aerosol species. Aerosol-forming vapors are important
40 from both an air quality and climate perspective. The nucleated aerosol particles can contribute to haze and reduced visibility, posing risks to both urban and rural environments, and alter Earth's radiation budget by scattering and absorbing sunlight, and acting as cloud condensation nuclei, which can alter cloud properties and precipitation patterns. Furthermore, trace gases are relevant for atmospheric chemistry in a number of ways, including in the formation and depletion of ozone, establishing the atmospheric oxidative capacity, and the oxidation of mercury, a potent neurotoxin (Khalizov et al., 2020; Shah et al., 2021).
45 Understanding the formation of short-lived reactive gases, and condensable vapors that can form aerosols is thus essential for addressing challenges in public health, air quality, and climate change.

Aircraft-based measurements are necessary and valuable for measuring atmospheric composition directly, and provide the vertical structure of constituents from the surface into the upper troposphere and lower stratosphere (UTLS) with a high
50 temporal and spatial resolution (Brenninkmeijer et al., 1999; Filges et al., 2015; Karion et al., 2010). These measurements complement and inform remote sensing observations on global scales, provide process level insights into atmospheric chemistry and physics, and are useful to assess predictions from chemical transport models about air quality and climate.

The aircraft gas inlet is one of the key components of an aircraft-based sampling system. It serves as the interface between the
55 ambient atmosphere and the analytical instruments onboard the aircraft, facilitating the collection of representative samples for analysis. Despite the advanced knowledge of measurement instruments (Kulkarni et al. 2011, Clemitshaw 2004), particle sampling methodologies (Kulkarni et al., 2011; Von der Weiden et al., 2009; Yang, 2017) and particle sampling inlets (Craig et al., 2014; Dhaniyala et al., 2003; Eddy et al., 2006; Moharreri et al., 2014) , the studies of gas sampling inlet (Fahey et al., 1989; Kondo et al., 1997; Ryerson et al., 1999; Yang et al., 2024) and gas transportation process are limited. More specifically,
60 there is a lack of understanding of the gas sampling loss of condensable vapors. The gas sampling loss can occur due to the inlet design, the properties of gas-phase species, and the operating conditions. Loss of condensable vapors can result in uncertainties in the measured concentrations, thereby reducing the accuracy and reliability of atmospheric data obtained from airborne platforms.

65 Understanding and quantifying gas sampling efficiency is essential for improving the accuracy of atmospheric measurements and enhancing our understanding of atmospheric processes. As the gas sample moves through the sample tube, gas-phase diffusion will drive gas-species in the sampled flow from a high concentration region to a low concentration region (Bird et al., 2006). For studying gas-phase diffusion in complex geometries, computational fluid dynamics (CFD) simulations have been used, and their performance validated in pipe-like geometries (De Schepper et al. 2008; 2009; Deendarlianto et al. 2016; 70 López et al. 2016). Applications of CFD method on modeling diffusion in gas-liquid, gas-solid and other two-phase flow regions are well applied and discussed (Hassanzadeh et al. 2009; Xin et al. 2015). Fick's law as a fundamental model of diffusion in mass transport is well studied for binary diffusion (Bird et al. 2006). As studies of diffusion are relevant to many different fields, Fick's law has been revised and adjusted for different applications (Lowney and Larrabee 1980; Van De Steene and Verplancke 2006). For our study, as demonstrated in part 1 of our inlet study (Yang et al., 2024), the prediction of turbulent 75 diffusivity has a significant impact on the mass transport model applied in the aircraft inlet. Yang et al. (2024) establishes that using the correct flow model is a critical pre-requisite for accurately predicting gas loss. Consistent with our previous study of modelling water vapor transport in the aircraft inlet, the same mass transport model is used to parametrically characterize gas phase H_2SO_4 loss in our sampling system.

80 In this paper, we aim to investigate the mass diffusion loss associated with our aircraft-based gas sampling inlet and sampling line. Using a combination of CFD simulations and gas-phase H_2SO_4 measurements in a high-speed wind tunnel, we seek to elucidate the relationship between gas sampling loss and sampling conditions, and ultimately provide the strategies to optimize the gas sampling efficiency through our aircraft-based sampling system. Section 2 describes our methodologies to study the gas-phase H_2SO_4 transmission through our sampling system. This includes both the experimental setup to measure gas-phase 85 H_2SO_4 transmission in the wind tunnel, and the procedure of computational simulations. Section 3 presents and discusses our experimental and CFD model results, the comparisons to evaluate the simulation data using the experiments, and the predictions of gas-phase H_2SO_4 sampling efficiency through our sampling system. Finally, section 4 summarizes our findings and presents conclusions as well as an outlook.

2 Method

90 The complete aircraft gas-sampling system includes: the aircraft gas inlet described as part of Yang et al. (2024), the aircraft sampling line, and a NO_3^- API-LToF-CIMS (Atmospheric Pressure Interface Long Time of Flight Chemical Ionization Mass Spectrometer with nitrate reagent ion) instrument. The nitrate ToF-CIMS was successfully utilized to measure gas-phase sulfuric acid (H_2SO_4), hydroxyl radicals (OH), and various other acids (Eisele and Tanner, 1991, 1993; Tanner et al., 1997). This technique has proven effective in providing accurate and sensitive measurements of these critical atmospheric 95 components. Our inlet design is based on the design by Eisele et al. (1997). It incorporates two shrouds to isolate and straighten the sample flow, with the sampling tube positioned at the center of the inner shroud to collect the sample flow without wall

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losses from the shrouds. The back of the inlet features a restrictor to slow down and control the flow rate. This design can facilitate in situ calibration of OH , H_2SO_4 , and other species, such as iodic acid (Eisele et al., 1997; Finkenzeller et al., 2023; Mauldin et al., 1998). Gas-phase H_2SO_4 is measured through the sampling system under different experimental configurations of the inlet (sampling line, restrictor size); and a range of different wind conditions and sampling flow rates were tested. The CFD flow and mass transport simulations represent the sampling system, boundary conditions and sampling flow rates. Both experimental results and simulation results were further analyzed to derive and compare the gas transmission efficiency.

2.1 Wind tunnel experimental setup

The sampling system of gas-phase H_2SO_4 measurements is shown in Figure 1. The inlet is installed inside a wind tunnel test section that is 0.3m by 0.3m in dimensions (Fig. 1a). The wind tunnel is operated at different freestream velocities (30, 75, 130 and 180 $m\ s^{-1}$) that represent the range of aircraft speeds. This generates the different external and internal flows of the inlet. The flow from the sample tube was further sub-sampled to transfer H_2SO_4 through a sampling line to the CIMS instrument (Fig. 1b). The sampling tube shown in Figure 1b is the actual sampling line used in-flight onboard the NSF/NCAR GV aircraft; it consists of a 33" (0.84 m) long line, made of stainless steel, with several bending sections built to fit the requirements of installation on the aircraft. The CIMS instrument was located directly outside the wind tunnel testing section (Fig. 1c) and connected via the sampling line. The CIMS measured real-time chemical signal in response to the changes of wind tunnel conditions and sampling conditions. A mass flow controller (MFC) is used to throttle a pump to maintain and vary the sampling mass flow rate (see Table 1). In addition, to evaluate the role of shape of the sampling line on the gas-phase H_2SO_4 transmission efficiency, three other types of sampling lines are also tested. The schematic diagram of the tested lines is shown in Figure 1d. The in-flight sampling tube with bends is labeled as type 1. The sampling tube labeled type 0 is a 33" (~0.84m) straight tube of the same total length as the in-flight tube (i.e. type 1). The type 2 and type 3 tubes are the 40" (~1m) straight tubes connected at the end of 33" (~0.84m) straight tube with and without 90° bend at the end, respectively. We note that the wind tunnel setup has an extra 90 degree bend compared to actual in-flight sampling system. This extra bend is identical in all sampling lines and thus cancels out in relative comparisons of transmission between different line type configurations. Since the overall inlet transmission for the in-flight sampling system relies on piecing together the individual segments of the inlet assembly as shown in Figure 5, the extra bend is not counted towards assessments of the aircraft configuration.

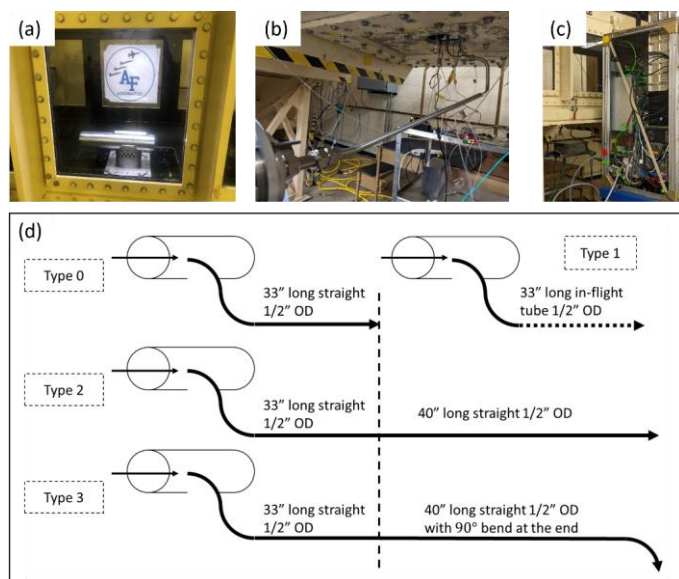
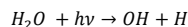


Figure 1. The wind tunnel experimental setup for H_2SO_4 transmission measurements. (a) Laminar gas inlet installed inside the wind tunnel test section. (b) The sampling lines used to connect the inlet with the CIMS instrument (c). The CIMS measured H_2SO_4 produced from the photolysis of H_2O at the tip of sampling tube inside the inlet under different line configurations, and wind tunnel conditions. (d) The schematic of four tested types of sampling lines; the line used in-flight is shown in (b) and referred to as type 1. The vertical dashed line marks the first section, which is the same length for each setup. For Type 1, the dashed line represents the different shape, but same length as Type 0.

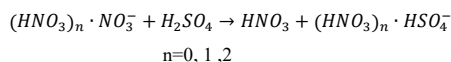
2.2 Wind tunnel experiments

To conduct gas-phase H_2SO_4 measurement, additional supporting devices are integrated into the inlet (Fig. 2). As shown in Figure 2, a UV source consisting of a Pen-Ray Mercury Lamp (90-0012-01), illuminates the inner shroud ~ 10 mm in front of the sampling tube entrance. Ambient water vapor from the free stream is photolyzed as it travels into the inlet through the outer shroud. The photochemical dissociation (photodecomposition) of water vapor upon interaction with the UV light generates hydroxyl radicals.



The center of the inner shroud flow is sub-sampled by the sampling tube. SO_2 is injected (~ 1 ppm tank mixture) via a port located near the entrance of the sampling tube. The hydroxyl radical interacts with SO_2 , oxygen and H_2O to generate H_2SO_4 (Finlayson-Pitts and Jr, 1999; Kolb et al., 1994). To prevent secondary radical cycling in the sampling tube, a small flow of Propane (~ 100 ppmv) is added downstream. The sampled gas containing H_2SO_4 continues through the sampling tube until it reaches the entrance to the CIMS IMR. The nitrate CIMS used here has been described in detail in many previous publications

(Eisele and Tanner, 1993; Mauldin et al., 1998), thus only a brief description will be given here. A schematic of the nitrate IMR is shown in Figure 2c. The sample flow enters the IMR via the sample tube as shown. Inside the IMR, surrounding the sample tube is a flow of filtered air that has been “spiked” with a small amount of HNO_3 . The outer annular region of this flow passes over an x-ray ionization source where the HNO_3 is ionized to form NO_3^- . Exiting the ionization region, the IMR flow consists of a central sample flow core, an annular region of unionized air surrounded by an outer region containing NO_3^- ions. The NO_3^- ions, but not the gas containing them, are then directed by means of electrostatic lenses into the central sample flow core where they interact with H_2SO_4 to form HSO_4^- via:



The ions are then directed by electrostatic lenses to the pinhole entrance of the Time of Flight mass spectrometer while the unionized gas is pumped away. The concentration of H_2SO_4 is then proportional to the ratio of the product ion signal, $(HNO_3)_n \cdot HSO_4^-$, to the reagent ion signal, $(HNO_3)_n \cdot NO_3^-$ (where $n = 0, 1, 2$). To account for processes that produce H_2SO_4 other than $SO_2 + OH$, measurements were also made where propane, an OH scavenger, was added along with the SO_2 . The product ion to reagent ion ratio from these measurements were subtracted from those without propane added to obtain the ratio due H_2SO_4 resulting from $SO_2 + OH$ only.

In addition, to emulate the different operating conditions and to examine the impact from different system designs, four parameters were varied in the wind tunnel experiments, i.e., restrictor size of the aircraft inlet, free stream velocities (U_∞), sampling line type, and sampling flow rate. The testing cases are summarized in table 1.

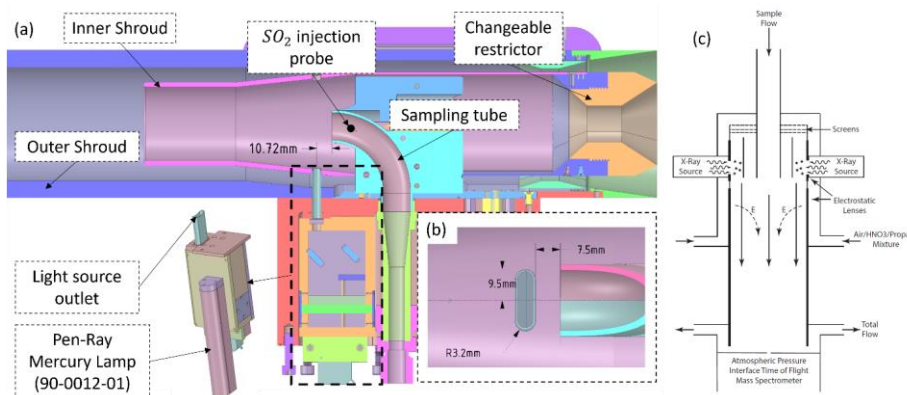


Figure 2. Cross-section view of the laminar gas inlet with UV lamp. (a) cross-section side view: shows different components of the inlet, incl. a 3d view of the UV source with mirror housing. (b) Cross-section top view: shows a detail view of the UV source outlet placement in front of the sampling tube entrance with dimensions. (c) CIMS Instrument Diagram.

Table 1. Variables of the wind tunnel H_2SO_4 gas measurements.

Restrictor size (mm)	12.5								17											
U_∞ ($m s^{-1}$)	30		75		130		180		30		75		130		180					
Sampling line type	0	1	0	1	0	1	0	1	0	1	2	3	0	1	2	3	0	1	0	1
Sampling flow rate (SLPM)	4, 8, 10	4, 8	4, 8, 10	4, 8	4, 8, 10	4, 8	4, 8, 10	4, 8	4, 8, 16	4, 8	4, 8, 16	4, 8, 16	4, 8, 16	4, 8	4, 8	4, 8, 16	4, 8	4, 8	4, 8	4, 8

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2.3 Computational flow and mass transport modelling for sampling line

To inspect the correlations between internal flow features and gas measurement results, the CFD simulation results from previous inlet studies were used. The details of the aircraft inlet CFD modelling was described in our first laminar gas inlet paper (Yang et al., 2024). Compared to the freestream conditions considered in the first paper, since gas-phase H_2SO_4 measurements were also conducted at freestream velocity of $30 m s^{-1}$; in gas measurement experiments, this freestream condition was added into our aircraft inlet simulations for both 12.5mm and 17mm restrictors. These additional simulations help facilitate direct comparisons with experimental results.

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To predict and compare the gas transmission efficiency with measurements, we conduct flow and mass transport modeling in different sampling tube designs using the commercial code FLUENT 18.1 (ANSYS, NH), focusing on a 40-inch (~1 m) long straight tube with an ID of 10.7 mm. For the flow modelling, as most sampling flow rates (Tab. 1) result in a Reynolds number less than 2300 in the tube, the laminar flow model is used. In addition, considering the case when Reynolds number is close to 2300, a small disturbance in the tube can cause turbulent diffusion loss, we also used the transition SST $k-\omega$ model for flow modelling across both laminar and turbulent flow regimes ($400 < Re < 7000+$). Both flow models were used to examine the influence of diffusion coefficient in mass transport with and without any turbulence effect. Due to the symmetric nature of the tube, both a 2D-axisymmetric solver and a 3D solver were tested. The mesh is refined at the near wall region by applying bias mesh gradient for the 2D model and inflation method for the 3D model. The final mesh size was selected such that any further refinement resulted in a less than 1% change in the flow velocity profile and mass fraction profile at the outlet of the tube. The final CFD model with a 3D geometry used ~1.6 million cells to simulate flow under different boundary conditions.

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To model gas-phase mass transport, we consider the role of mass diffusion due to the concentration difference of gas-phase H_2SO_4 between the sample flow and the tube wall. In addition, we consider the diffusion process as binary diffusion, i.e., the model calculates gas-phase H_2SO_4 diffusion in the dry air. The binary diffusivity of H_2SO_4 in the laminar flow regime, referred as H_2SO_4 laminar diffusivity, is set as constant value of $1.04e^{-5} m^2 s^{-1}$. The Fick's law of diffusion is well applied for binary diffusion, and the model is applied in FLUENT 18.1. In addition, as the temperature gradient in the transmission line is insignificant, we neglect thermal diffusion loss. The simplified governing equation of our mass transport model expressed as:

$$\frac{\partial \rho \bar{w}_i}{\partial t} + \nabla \cdot (\rho \bar{v} \bar{w}_i) = -\nabla \cdot \vec{j}_i$$

where the mass fraction w_i represents the relative density of the diffusive species ρ_i ($kg m^{-3}$) to the mixture density ρ ($kg m^{-3}$), i.e. $w_i = \frac{\rho_i}{\rho}$. The v ($m s^{-1}$) represents the flow velocity. The expressions of mass flux j_i is determined by

different flow models.

$$j_i = -\rho D_{ij} \nabla \bar{w}_i ; \text{laminar flow model}$$

$$j_i = -(\rho D_{ij} + \rho D_t) \nabla \bar{w}_i , \text{turbulent flow model}$$

where D_{ij} ($m^2 s^{-1}$) represents the laminar diffusivity, the subscript i represents the studied species (gas-phase H_2SO_4 or water vapor), the subscript j in our study represents dry air, and D_t ($m^2 s^{-1}$) represents the turbulent diffusivity.

For the laminar flow model, the laminar diffusivity D_{ij} is a constant value and only determined by the diffusing species, here gas-phase H_2SO_4 . For the turbulent flow model, both laminar and turbulent diffusion contribute to the mass flux. The turbulent diffusivity D_t is calculated based on the turbulent viscosity predicted from the turbulent flow model, $D_t = \frac{\mu_t}{SC_t}$, where SC_t [turbulent Schmidt number is dimensionless parameter that describe diffusion in mass transfer](#) (Kang and Yang, 2014).

The different sampling conditions are modelled to investigate the factors that influence the gas transmission efficiency. The sampling flow rate and the mass fraction of H_2SO_4 on the wall are two key factors we investigated. [The wall mass fraction boundary condition determines the probability that a gas molecule in the flow is captured on the wall. With a zero mass fraction boundary condition, the wall acts as a perfect sink and every gas molecule is accommodated on the wall. A wall mass fraction of one implies the wall maintains a layer of the gas on it, and the wall effectively does not accommodate any molecules from the flow. Boundary conditions between these two extreme values results in varying ability of the wall to accommodate gas molecules from the flow. Thus, the ratio of species mass fraction on the wall to that in the flow can be related to an equivalent wall accommodation coefficient \(\$\alpha_i\$ \) for a selected gas species, calculated as:](#)

$$\alpha_i = 1 - \frac{w_{wall}}{w_{flow}}$$

[And different mass accommodation coefficients, \$\alpha_i\$, were used to represent different boundary conditions on the wall for mass transport modelling. Different laminar diffusivities are used to represent different species or operating conditions. The entire set of boundary conditions for flow modeling and tested laminar diffusivities are shown in Table 2. The mass accommodation](#)

coefficient is defined as number of gas molecules taken up by the surface divided by number of gas-surface collisions (Finlayson-Pitts and Jr, 1999). To simulate different mass accommodation coefficients, we set different species mass fraction boundary condition on the inlet walls. The equation expresses the relationship between mass accommodation coefficient and mass fraction as follows:

$$\alpha_i = 1 - \frac{w_{wall}}{w_{flow}} = \frac{w_{flow} - w_{wall}}{w_{flow}}$$

Using this approach, the mass accommodation coefficient essentially simulates different mass fluxes toward the wall and is effectively set between the limits of 0 and 1, depending on the species mass fraction on the walls. The influence of varying wall mass fraction boundary conditions on the species accommodation is discussed in our previous paper (Yang et al., 2024).

The varying boundary conditions of mass fraction of H_2SO_4 on the wall relates to varying α values. For our mass transport modeling, α represents the mass accommodation coefficient of a studied species diffused in the dry air. When $\alpha_i = 1$, the mass fraction of H_2SO_4 set on the wall is 0, the tube wall is a perfect sink and provide provides us the worst sampling ease with the maximum near wall mass flux. When α_i decreases, the mass fraction of H_2SO_4 set on the wall increase. When $\alpha_i = 0$, the mass fraction of H_2SO_4 on the wall has the same mass fraction of H_2SO_4 in the flow, and no species will be lost to the walls due to the absence of a mass concentration gradient. If $\alpha_i > 1$, the near wall mass flux takes a negative value, indicating that the species is released from the wall into the flow.

In our simulations, we vary the wall mass fractions to vary the effective wall mass accommodation coefficient. We vary the laminar or molecular diffusivities to represent different species or operating conditions. The entire set of boundary conditions for flow modeling are shown in Table 2. Moreover, the ambient operating conditions in the sampling line are obtained from aircraft inlet simulations under both ground level and high altitude conditions. All simulations were modelled assuming steady state flow.

Table 2. Boundary conditions for flow modelling in 40" straight sampling line.

Flow model	Laminar flow model	SST turbulent model
Sampling flow velocity ($m s^{-1}$)	0.6, 1.2, 2.4, 3.6	0.6, 1.2, 2.4, 3.6, 4.8, 6, 7.2, 8.4, 9.6, 10.8, 12, 13.2, 14.4
Mass accommodation coefficient (α_i)	1, 0.75, 0.65	1, 0.75, 0.65, 0.5, 0.25, 0
Mass diffusivity D_{ij} ($m^2 s^{-1}$)	1.04 x 10 ⁻⁵ , 2.88 x 10 ⁻⁵ , 4.5 x 10 ⁻⁵	

3. Results and discussions

The CIMS measurement data was systematically analysed with different experimental setups, operating conditions, and time periods. We calculate uncertainties using propagation analysis and the detailed error analysis is shown in the supplement. The CFD simulation results are used to compare and explain the observations from experimental results. The following sections will illustrate the data process and our findings.

3.1 Wind tunnel measurements

The transport efficiency measurements were made by analyzing the transport of species HSO_4^- and $HNO_3 \cdot HSO_4^-$ through the different sampling line types. These ion concentrations were recorded under different operating conditions by CIMS instrument (Fig.1 and Tab.1). Although applying the calibration factor of H_2SO_4 can convert these relative measurements to concentrations, the comparison between the sampling line types only relies on the use of relative information, and thus cancels out the calibration factor for sulfuric acid without adding uncertainty of calibration factor. As Figure 3a is shown, the wind tunnel maintains the freestream velocity at different speeds for a measurement period and thus emulates different flight conditions for the gas-phase H_2SO_4 measurement. The temperature of the wind tunnel is increasing the longer the tunnel operates. Increases in temperature can provide slight variations in relative humidity, while the absolute humidity inside the wind tunnel is set by the outside air which re-circulates and cools the wind tunnel. The measurement of gas-phase sulfuric acid H_2SO_4 is presented as the sum of HSO_4^- and $HNO_3 \cdot HSO_4^-$ divided by the sum of nitrate parent ions (H_2SO_4 NCPS).

$$\frac{H_2SO_4}{\sum_{n=0}^2 (HNO_3)_n \cdot NO_3^-} \cong \frac{HSO_4^- + HNO_3 \cdot HSO_4^-}{\sum_{n=0}^2 (HNO_3)_n \cdot NO_3^-}$$

This ion ratio signal is unitless, and referred to as normalized counts (NCPS) or reagent normalized signal. A typical calibration factor for sulfuric acid is 5×10^9 molec cm^{-3} for the source configuration used (Eisele and Tanner, 1993; Mauldin et al., 1998); NCPS multiplied by the calibration factor yields a sulfuric acid concentration ($1-2 \times 10^7$ molec cm^{-3}) generated at the tip of the sampling tube in the aircraft inlet (Fig. 2a). In practice, the amount of sheath and total sampling flow were adjusted to give good NCPS signal, and the calibration factor is a function of the sample flow rate. However, the exact value of the calibration factor, and its uncertainty, does not affect the results, since our further analysis only relies on the ratio of NCPS values under comparable sample flow rates. The value of the calibration factor hence cancels out in relative statements about inlet transmission. For each measurement period, the CIMS instrument repeated three measurements of the raw data of H_2SO_4 NCPS with both background mode and signal mode (Fig. 3b). The background mode measures gas-phase H_2SO_4 background signal that is generated from dark sources, without addition of UV light. This mode provides the baseline of H_2SO_4 measurement under each operating condition. The signal mode provides the gas-phase H_2SO_4 measurement with additional hydroxyl radical, generated by photochemical decomposition of water vapor when sample air passes the UV light source. The signal depends on the operating conditions; i.e., NCPS of sulfuric acid decreases as the free stream velocity is increased.

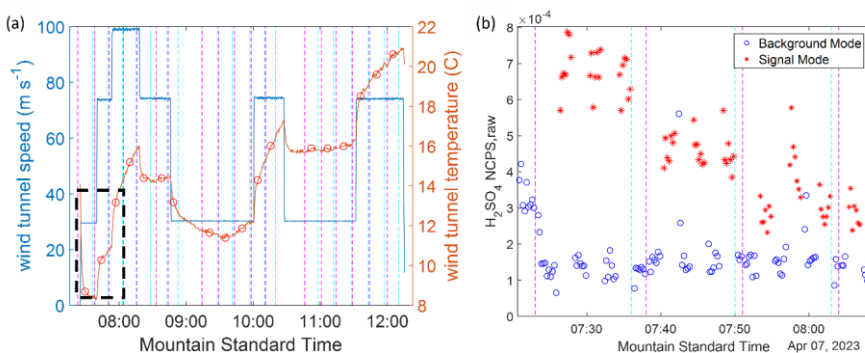


Figure 3. Wind tunnel conditions for experiments on Apr 07, 2023. (a) (blue line) free stream velocity inside the wind tunnel; (red line) temperature inside the wind tunnel. (black circles) averages for the gas measurement periods. (b) Real time H_2SO_4 NCPS raw measurements for the first three measurement periods marked in (a). (blue circles) background mode: detect hydroxyl radical pre-existing in the sample air; (red stars) signal mode: detect hydroxyl radical come from both sample air and water vapor photochemical decomposition. The magenta and cyan dashed lines for both figures mark the beginning and the end of each measurement case, respectively.

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The difference between signal mode and background mode is proportional to the gas-phase H_2SO_4 concentration that is generated by the water vapor photolysis in the sample air (H_2SO_4 NCPS). Therefore, the measurement result is naturally dependent on the water vapor concentration in the ambient air. As the wind tunnel experiment was completed over multiple days, the repeated measurements under the same operating condition and setup shows significant differences that are the result of the changes in specific humidity under different weather conditions on different experiment days (Fig. S1). These differences result in varying specific humidity inside the wind tunnel (Fig. S1a), and needed to be accounted for in comparing the measurement results of H_2SO_4 NCPS between different experiment days. A correction factor that normalizes changes in specific humidity (f_q) was defined as specific humidity (q) at each measurement period divided by the mean value of specific humidity among all measurements, and is found to largely reduce the differences among repeated measurements (Fig S1b). The measurements of H_2SO_4 NCPS were normalized for constant specific humidity, H_2SO_4 NCPS/ f_q , for further analysis and comparisons. The detailed analysis for raw data and correlation between flow and experimental results, H_2SO_4 NCPS/ f_q , is shown in supplement.

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3.2. Gas transmission efficiency comparisons

All types of sampling lines (Fig. 1d) and the different sampling flow rates are tested for gas-phase H_2SO_4 measurements using the 17mm restrictor (Tab. 1). The normalized measurement results (H_2SO_4 NCPS/ f_q) from 30 m s⁻¹ freestream velocity is shown in Figure S5. The type 0 transmission line, due to the shortest length and minimum bends, shows the highest signal under comparable sampling flow rates, compared to other types of sampling lines. Increasing the length of the sampling line

295 decreases the measured signal, consistent with additional losses in longer tubes. In addition, the 8 SLPM sampling flow rate maintains the highest quantity of signal, while 16 SLPM shows the lowest signal among three different sampling flow rates. This is specific to the instrument configuration used here, and related to the reaction time and flow characteristics inside the ion molecule reaction chamber of the CIMS instrument. Further analysis is outside the scope of this paper, which compares only ratios of normalized signal at comparable sampling flow rates. Similar trends are also observed in all cases of 75 m s^{-1} freestream velocity. We are choosing the cases from the 30 m s^{-1} freestream velocity for further analysis of the gas transmission efficiency in the sampling line due to the higher signal-to-noise. It should be noted that upstream effects (f_q and f_{RT}) are accounted for in the normalization of the signal, ensuring that the results for transmission are applicable to other free stream flow cases and restrictors of aircraft inlet as well. For comparisons with the same restrictor, f_{RT} cancels out.

305 The gas transmission efficiency is defined as the mass fraction (or concentration) of a molecule (here: H_2SO_4 or H_2O) at any cross-section along the sampling pathway compared to a reference mass fraction. Based on the wind tunnel experiments, we calculated the gas transmission efficiency of gas-phase H_2SO_4 measurement in the 40" (1 m) tube as the ratio of normalized signal ($H_2SO_4 \text{ NCPS}/f_q$) from the type 2 and type 3 sampling lines divided by the type 0, respectively, under each same sampling flow rate (f_{RT} cancels out). We use the mass fraction of the simulated H_2SO_4 at the outlet of the 40" straight tube divided by its initial value at the tube inlet to calculate the gas sampling efficiency under different boundary conditions in the model. The experimental and simulated transmission are compared in Figure 4. The experimental results clearly show the gas transmission efficiency in the 40" tube is far below 1 for all sampling flow rates, which indicates a gas loss in this tube. In addition, the experimental results also show an increasing trend of the transmission efficiency as the sampling flow rate is increased. Moreover, the simulation results over predict the observed loss under the assumption that the mass fraction of H_2SO_4 is 0 on the wall, which corresponds to a mass accommodation coefficient $\alpha_i = 1$. Both the laminar flow model and the SST turbulent model have good agreement with the experimental results for accommodation coefficients between 0.65 and 0.75 for gas-phase H_2SO_4 . This agreement not only provides model verifications of the conclusion that reducing the residence time during the gas-phase species transport can reduce the gas transmission loss, but also indicates the accommodation coefficient of H_2SO_4 is around 65% to 75% which agrees with [related findings in](#) other studies (Hanson, 2005; Pöschl et al., 1998). In Pöschl's paper, the mass accommodation for H_2SO_4 at 303K under dry conditions ($\text{RH} \leq 3\%$) has a best fit value of 0.65, with a physical upper limit of 1, and a lower statistical limit of 0.43. Hanson further discussed the impact of RH on the mass accommodation coefficient, and their data for H_2SO_4 show an average value of 0.76 for $\text{RH} < 50\%$. Our experimental results [align](#) reasonably ~~align~~ closely with both previous laboratory studies, and the simulation results that apply a mass accommodation coefficient for H_2SO_4 of 0.65 and 0.75 best describe the experimental observations in our aircraft inlet line.

325 Therefore, we selected 0.7 as the average mass accommodation coefficient and use $\pm 5\%$ as uncertainty for further simulations. Moreover, the SST turbulent model overlaps with the laminar flow model when the flow rate is low and deviates when flow is outside the laminar flow regime. This observation provides further confidence in predicting the turbulent effects in our

330 sampling system using the SST model, consistent with the findings comparing hotwire measurements of turbulence described in part 1 of our inlet system analysis (Yang et al., 2024). Overall, the flow and mass transport models provide a reasonable description of the gas transmission loss in the entire inlet sampling system.

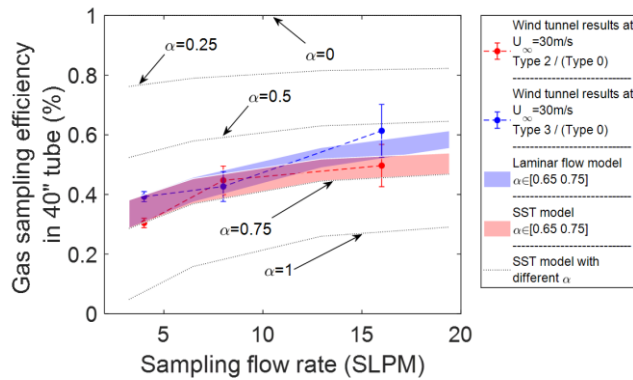


Figure 4. Comparison of different types of sampling tubes and the gas transmission efficiency: Comparison of the gas sampling efficiency measured along 40" (~1 m) of a straight sampling tube with simulation results that vary the accommodation coefficient, α_i . The error bar is uncertainty of the data which follows the description of propagation analysis in the supplement.

335 3.3 Overall gas sampling efficiency of H_2SO_4

The verified CFD model assumed 70% for the mass accommodation coefficient α_i to set the boundary condition on the surface wall for gas-phase H_2SO_4 transport modelling. The gas-phase H_2SO_4 transmission efficiency of the overall inlet sampling system is calculated as a product of the transmission in different sampling sections. Specifically, Figure 5 illustrates the individual components used to assess the overall gas-phase H_2SO_4 transmission efficiency: (a) transmission through the 12.5mm restrictor inlet; (b) a straight 33" (~0.84m) tube; (c) losses due to the shape factor (type 0 compared to type 1) due to the bending features (complements losses under (b)) with 10% uncertainty; and (d) a straight 20" (0.5m) tube, respectively. The overall uncertainty in the transmission of the aircraft inlet sampling system combines the uncertainty in an ideal sampling system under various flow rates and flight conditions with a $\pm 5\%$ variation of the selected mass accommodation coefficient, and the shape factor ($\pm 10\%$). In addition, to predict sampling efficiency for high altitude, we choose $4.5 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ for the laminar diffusivity. This value is calculated based on the pressure and temperature at high altitude condition refer to the laminar diffusivity of gas-phase H_2SO_4 at ground level, $D_{ij} = D_0 \left(\frac{T}{T_0}\right)^{1.75} \frac{p_0}{p}$, where the variables with 0 substitute present the reference value.

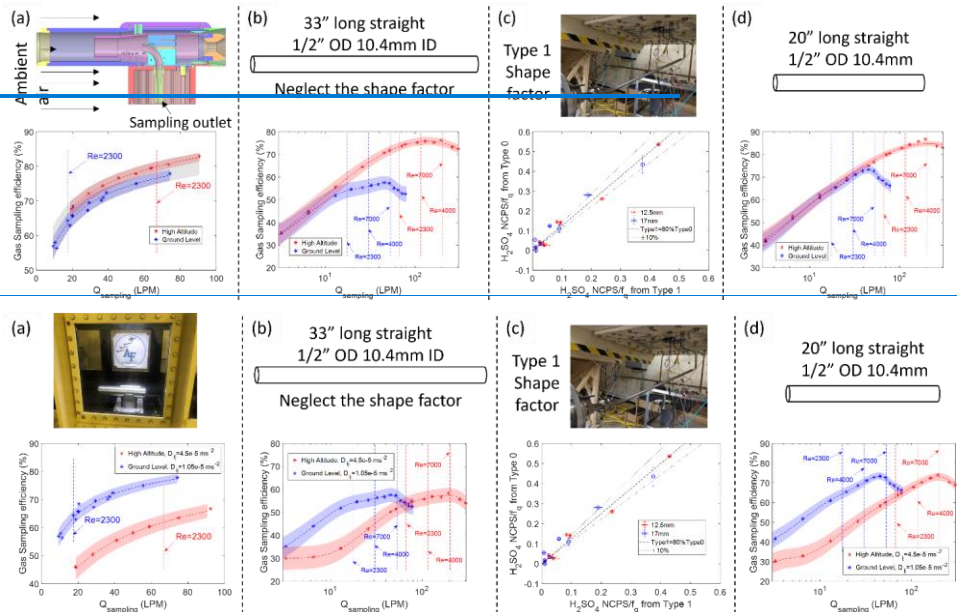
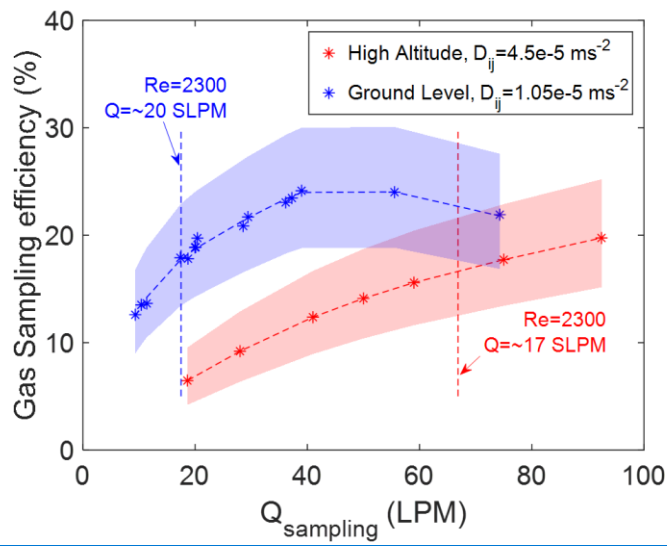
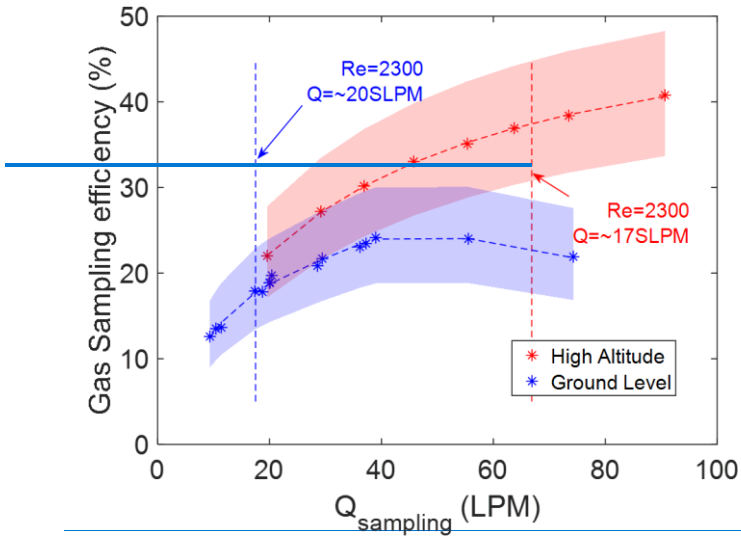


Figure 5. The gas-phase H_2SO_4 sampling efficiency in each partial section of the in-flight sampling system. The shaded areas in (a), (b) and (d) presents $\pm 5\%$ uncertainty of the selected mass accommodation coefficient (70%); the overall sampling efficiency is shown in Figure 6. Panel (5a) Inlet section: ambient air to the sampling outlet; (b) 33" (0.84m) straight tube; (c) Shape factor: compares type 1 and type 0 under same operating conditions; (dashed line) fitted as $Type\ 1 = 80\% \times Type\ 0$; (dotted lines) $\pm 10\%$ uncertainty of shape factor; (d) 20" (0.5m) straight tube to connect the CIMS in the aircraft configuration.

Notably, the individual sampling sections influence the gas-phase H_2SO_4 transmission efficiency differently. For the aircraft inlet section, the sample flow in the gas inlet is mainly impacted by the upstream turbulence. As a result, the gas sampling efficiency through this section shows a direct correlation with the sampling flow rate, and a negligible difference at the two flight altitudes investigated. is mainly due to the laminar diffusivity at high altitude flight condition is set to be ~5 times larger than ground level (Fig. 5.a). Similarly, However, since the sample flow in the sampling line is directly impacted by the characteristics of the local flow, the prediction of gas transmission efficiency inside the sampling line is significantly also different between ground level and high altitude (Fig. 5b & 5d). At ground level, due to the higher flow density, the flow in the sampling tube is reaching the turbulent regime at a lower volume flow rate than at high altitude. In addition, the simulation results from both altitudes consistently show that the gas sampling efficiency can increase even when the sample flow enters the turbulent flow regime ($Re > 4000$). However, when flow reaches higher turbulent ($Re > 7000$), the sampling efficiency in sampling tube starts dropping again. This observation is due to the higher prediction of turbulent diffusivity, and indicates

that the fate of the overall inlet gas transmission efficiency is determined by the local sampling flow rate (Q_{sampling}). Since
For a selected sampling flow rate Q_{sampling} , the Reynolds Number in the tube can be expressed as $Re = \frac{4Q\rho}{D\pi\mu}$, at a fixed
sampling flow rate Q_{sampling} , the results imply that enlarging-increasing the inner diameter of the sampling tube (D) can
370 reduces the Reynolds number ($Re = \frac{4Q\rho}{D\pi\mu}$), and thus correspondingly reduce the modeled prediction of turbulence in the
sampling tube, with benefits for the overall gas transmission efficiency. In order to achieve the maximum gas transmission
efficiency, our simulation results suggest that the optimized flow rate should maintain the Re in the sampling tube at ~6000.

The overall gas sampling efficiency of the actual in-flight sampling system is calculated by multiplying each the sampling
375 efficiency at the different sections and shape factors together. The results are shown in Figure 6. We present the overall
sampling efficiency of the actual in-flight sampling system at two altitudes as a function of the sampling flow rate. The shaded
areas in Figure 6 represent the largest range of uncertainty from $\pm 5\%$ variation of mass accommodation coefficient and $\pm 10\%$
variation of shape factor. The simulation results indicate the overall gas sampling efficiency is increasing as the with sampling
flow rate is increased at both altitudes, and for most sampling conditions. This effect extends even when the sampling flow
380 result in a Reynolds number is pasting the laminar flow regime ($Re \leq 2300$); These simulation results and reflects suggest
that the residence time inside the inlet determines overall inlet transmission even well into the turbulent flow regime. However,
the turbulent diffusivity predicted from the flow model increases as the Reynolds number increases. The overall sampling
efficiency increases less rapidly, and eventually starts to decrease when the sampling flow rate reaches a much higher Reynolds
number level ($Re > 7000$). The overall sampling efficiency is lower at ground-level high altitude compared to high
385 altitude ground level if the results are compared at the same flow rate. This is due because to sample flow is more turbulent at
lower altitudes the laminar diffusivity at high altitude is 5 times more than the value set at ground level. Although the low
altitude produces more turbulence and generates higher turbulent diffusivity, the significant change of laminar diffusivity
also plays an important role in the sampling efficiency calculation. Overall, based on the simulation results, suggest that
the overall sampling efficiency of gas-phase H_2SO_4 for the actual in-flight sampling system can increase by a factor of 2 (from
390 12% @ 10 SLPM sampling flow rate to ~20% @ ~30 SLPM) by increasing the sampling flow rate at ground level. At high
altitude, due to lower air density operating pressure resulting into in a lower much higher laminar diffusivity, the overall
sampling efficiency can increase to ~40% by increasing the sampling flow rate.



395 Figure 6. The correlation between overall gas transmission efficiency of H_2SO_4 and sampling flow rate $Q_{sampling}$ (LPM) at ground level and high altitude. The dash lines mark the Reynolds Number in sampling line equal to 2300, which distinguish the laminar flow regime and beyond. The shade areas present the range of uncertainty from $\pm 5\%$ variation of mass accommodation coefficient and $\pm 10\%$ variation of shape factor.

400 3.5 Extending transmission to other species

The transmission efficiency of H_2SO_4 can be extended also to other species ~~or other operating conditions by directly changing the value of laminar diffusivity D_{ij} .~~ However, a detailed treatment of other species requires a careful consideration of ~~selecting the laminar~~ diffusivity and mass accommodation coefficients that is beyond the scope of this work. We ~~therefore limit discussion on other species to sensitivity studies in a straight tube that with varying laminar diffusivities (i.e., illustrate the effects using water vapor as an example molecule with $2.88 \times 10^{-5} (m^2 s^{-1})$ and $4.5 \times 10^{-5} (m^2 s^{-1})$ for higher laminar diffusivities)~~ ~~y,~~ and discuss some general considerations to inform future work. Simulations were performed for ~~water vapor using these a laminar~~ diffusivity of $2.88 \times 10^{-5} (m^2 s^{-1})$ values and varying the mass accommodation coefficient from 0.1 to 1. ~~The laminar or molecular diffusivity value of $2.88 \times 10^{-5} (m^2 s^{-1})$ was selected with respect to is consistent with gas-phase water vapor at ground level. The value of $4.5 \times 10^{-5} (m^2 s^{-1})$ was selected with respect to for the gas-phase H_2SO_4 at high~~ altitude flight condition. These values can vary depending on the species and operational conditions. In this study, we are not focusing on how to select laminar diffusivity values, but rather on showing the importance of choosing appropriate values.

The sample flow in the aircraft inlet section ~~and the laminar diffusivity is are~~ mainly impacted by the upstream flow conditions. ~~Thus However, the laminar diffusivity affects gas loss in a relatively short distance compared to the sampling section., Thus,~~ the gas sampling efficiency of the aircraft inlet section can be normalized for both altitudes and ~~both species different diffusivities~~ into a single fit function by using the volume flow rate divided by the laminar diffusivity, ~~which is effectively the species Peclet number as a new variable~~ (Fig. S6). As such, the gas-transmission efficiency for ~~this section aircraft of the inlet~~ system can be parameterized and solved analytically for different accommodation coefficient values for different species. However, no similar unified correlation of gas sampling efficiency can be found for ~~varying the sampling line between the two species among different diffusivities.~~ This is because gas loss is a result of a complex combination of flow velocity, flow turbulence, and laminar diffusivity ~~acting in over a long distance (~1.3 m).~~ In contrast, ~~the~~ laminar diffusivity plays a ~~smaller shorter secondary~~ role in the aircraft inlet section ~~(Fig. 5a),~~ owing to the larger, ~~dominating~~ role of turbulence in this section of the aircraft inlet system.

425 We ~~have~~ conducted a series of sensitivity studies to ~~illustrate understand the the gas species transmission for species with different diffusivity characteristics for through~~ a straight tube. Figure 7 presents the results of gas sampling efficiency in a 33" (0.84 m) straight tube between the ~~two species maximum~~ diffusivity of $4.5 \times 10^{-5} (m^2 s^{-1})$ and minimum diffusivity of 1.04

$\times 10^{-5} (m^2s^{-1})$ with different mass accommodation coefficients at both ground level and high altitude for future interpolations. Comparing the results of transmission efficiency for the two species studied, it is observed that the laminar (or molecular) diffusivity of a species plays a significant role in the prediction of the gas transmission efficiency when the mass accommodation is close to 1. However, this dependence on laminar diffusivity, however, decreases becomes negligible when the mass accommodation coefficient values is less than 0.25. The shaded area shown in Figure 7 (factor ~3-4 range higher in laminar diffusivity (shaded area) also corresponds to approximately illustrates provide perspectives that the magnitude of the combined effect of temperature and pressure changes between the low and high altitude cases that higher laminar diffusivity at lower pressure and temperature has on the gas transmission efficiency when sampling at higher altitude. For most tropospheric sampling, this the effect from mass accommodation losses coefficient will be minor for non-sticky molecules (mass accommodation < 0.25), but it gains in relative importance for sticky molecules sampled at low pressures; which was not the focus of the current study (windtunnel experiments at ground conditions), and deserves further attention in the future for high altitude sampling.

This paper emphasizes the importance of choosing the correct diffusivity and mass accommodation coefficient in predicting gas sampling efficiency of condensable vapors, in particular for highly diffusive condensable vapors (e.g., NH_3). Additionally, Khalizov et al. (2020) estimate that the limits of detection of oxidized mercury species are probably similar to those reported for H_2SO_4 (Eisele and Tanner, 1993; Mauldin et al., 1998; Zheng et al., 2010). Given the complexities of sampling oxidized mercury (Elgiar et al., 2024) significant transmission losses can be expected, with implications for the attainable detection limits in future attempts to sample oxidized mercury species using atmospheric pressure CIMS techniques (Khalizov et al., 2020). Moreover, the chemical properties of the inlet line can further influence transmission (e.g., pH, surface composition, reactivity, etc.). Determining these parameters and their transmission in flight warrants future work that is beyond the scope of this paper. The further warming of the sample in the tube as air transfers into the instrument aboard the aircraft depends on many parameters, including the temperature gradients towards along the tube as the flow enters the cabin air, the sampling flow rates, heat transfer from the tube to the gas, etc. These parameters and their influence on sampling efficiency are and this is currently not well characterized and cannot be generalized; and this-it is not easily possible to model either the full system accounting for all these parameters. We are planning to measure this gradient during an upcoming campaign to further characterize the flow conditions inside the sampling tube in flight.

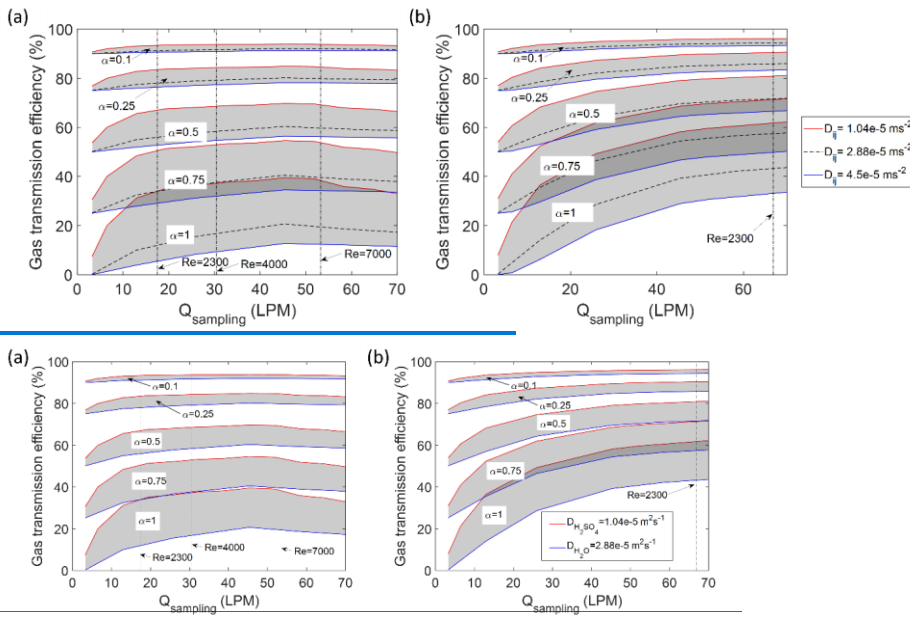


Figure 7. The comparisons of gas sampling efficiency between water vapor and gas-phase H_2SO_4 different laminar diffusivities at the sampling line section. (a) The correlation of gas sampling efficiency in 33" (0.84m) straight tube at ground level condition (1013 mbar). (b) The correlation of gas sampling efficiency in 33" (0.84m) straight tube at ground high altitude level condition (220 mbar). The red line shows the sampling efficiency of gas-phase H_2SO_4 with diffusivity set as $1.04 \times 10^{-5} m^2 s^{-1}$. The blue line shows the sampling efficiency of water vapor with diffusivity set as $2.8845 \times 10^{-5} m^2 s^{-1}$. The shade areas covered the predicted sampling efficiency between water vapor and gas-phase H_2SO_4 these diffusivities at a selected mass accommodation coefficient. The dark dash line in the shade areas shows the sampling efficiency with diffusivity set as $2.88 \times 10^{-5} (m^2 s^{-1})$.

4. Summary

In this paper, we describe an experimental approach to measure gas transmission efficiency of an aircraft inlet system using H_2SO_4 vapor and a high-speed wind tunnel setup. We use these observations to evaluate a CFD model of the aircraft inlet system; and use the validated CFD simulations to predict the overall sampling efficiency of gas-phase H_2SO_4 over a range of in-flight sampling conditions that span from the marine boundary layer (MBL) into the upper troposphere and lower stratosphere (UTLS). The inlet transmission for a condensable vapor like H_2SO_4 can be optimized using the sampling flow rate as a variable. For a realistic range of sampling flow rates, increasing the sampling flow rate can help cut transmission losses, and double the overall sampling efficiency of gas-phase H_2SO_4 compared to laminar flow conditions. The transmission

of H_2SO_4 can reach up to 40% in the UTLS, and up to 20% in the MBL, owing primarily to the lower turbulent intensity and shorter residence time of air inside the sampling line under conditions typical of the UTLS.

475 Experiments and simulation results consistently support the conclusion that increasing the sampling flow rate can increase the gas transmission efficiency. These results challenge the widely accepted assumption that laminar core sampling, i.e., sampling from the core flow inside of an inlet line operated under laminar flow conditions, minimizes wall losses. This assumption is currently very widely used in laboratory and field experiments that sample ambient trace gases and aerosol species. Instead, shortening the residence time that air spends inside inlet lines is found to minimize wall losses well into the turbulent flow regime. The maximum inlet transmission is found for $Re \sim 6000$, with lower transmission at both lower and higher Re numbers. 480 For a condensable vapor like H_2SO_4 , the effects to optimize transmission losses by optimizing the residence time can be as large as a factor of 2 over laminar core sampling.

To predict the transmission efficiency for other gas-phase species through this aircraft sampling system, the appropriate selection of the gas-phase diffusivity (D_{ij}) and mass accommodation coefficient (α_i) of the species (i) is necessary. The overall sampling efficiency is calculated by combining the transmission efficiencies of the aircraft inlet and sampling line. For the aircraft inlet, a simulation-based function predicts transmission efficiency based on D_{ij} and α_i , as the gas loss is affected only by the upstream conditions regardless of altitude. Other factors can further affect transmission, i.e., pressure and temperature dependencies of the laminar diffusion coefficient, and for reactive species multiphase chemistry in addition to physical factors need to be considered. ~~Due to the complexity and feasible~~With the computational cost of modeling gas loss in the straight sampling line ~~being reasonable~~, it is recommended to directly calculate the line's transmission efficiency using simulation models. Our flow analysis ~~illustrates with straight tubes of variable lengths and sampling conditions, illustrates~~ that the exact value of ~~laminar (molecular) diffusion, D_{ij} , (and their temperature and pressure dependencies)~~ only plays a role for sticky gases with $\alpha_i > 0.25$. ~~We provide simulation data for two species (gas phase H_2SO_4 and water vapor) three different laminar diffusivities under various tube lengths and sampling conditions for future interpolation in sampling lines.~~By combining the transmission efficiencies from all sections and incorporating the sampling line's shape factor, the overall sampling efficiency of a gas-phase species for this aircraft-based sampling system can be estimated. 495

Data availability:

The dataset is available at: [a zenodo link will be added here for publication](#)

500 **Supplement:**

The supplement related to this article is available online at:

Author contribution:

RV designed research. DY developed CFD model and conducted the simulations. RV, RM and EA carried out the wind tunnel experiments. DY analysed the data. DY, SD and RV wrote the manuscript, with contributions from all coauthors.

505

Competing interests:

At least one of the (co-)authors is a member of the editorial board of Atmospheric Measurement Techniques.

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