# Interferences on Aerosol Acidity Quantification due to Gas-phase Ammonia Uptake onto Acidic Sulfate Filter Samples

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**Abstract**

Measurements of the mass concentration and chemical speciation of aerosols are important to investigate their chemical and physical processing from near emission sources to the most remote regions of the atmosphere. A common method to analyze aerosols is to collect them onto filters and to analyze filters off-line; however, biases in some chemical components are possible due to changes in the accumulated particles during the handling of the samples. Any biases would impact the measured chemical composition, which in turn affects our understanding of numerous physico-chemical processes and aerosol radiative properties. We show, using filters collected onboard the NASA DC-8 and NSF C-130 during six different aircraft campaigns, a consistent, substantial difference in ammonium mass concentration and ammonium-to-anion ratios, when comparing the aerosols collected on filters versus the Aerodyne Aerosol Mass Spectrometer (AMS). Another *on-line* measurement is consistent with the AMS in showing that the aerosol has lower ammonium-to-anion ratios than obtained by the filters. Using a gas uptake model with literature values for accommodation coefficients, we show that for ambient ammonia mixing ratios greater than 10 ppbv, the time scale for ammonia reacting with acidic aerosol on filter substrates is less than 30 s (typical filter handling time in the aircraft) for typical aerosol volume distributions. Measurements of gas-phase ammonia inside the cabin of the DC-8 show ammonia mixing ratios of 45±20 ppbv, consistent with mixing ratios observed in other indoor environments. This analysis enables guidelines for filter handling to reduce ammonia uptake. Finally, a more meaningful limit-of-detection for SAGA filters collected during airborne campaigns is ~0.2 μg $sm^{-3}$ ammonium, which is substantially higher than the limit-of-detection of the ion chromatography. A similar analysis should be conducted for filters that collect inorganic aerosol and do not have ammonia scrubbers and/or are handled in the presence of human ammonia emissions.

## Introduction

Particulate matter (PM), or aerosol, impacts human health, ecosystem health, visibility, climate, cloud formation and lifetime, and atmospheric chemistry (Meskhidze et al., 2003; Abbatt et al., 2006; Seinfeld. and Pandis, 2006; Jimenez et al., 2009; Myhre et al., 2013; Cohen et al., 2017; Hodzic and Duvel, 2018; Heald and Kroll, 2020; Pye et al., 2020). Quantitative measurements of the chemical composition and aerosol mass concentration are necessary to understand these impacts and to constrain and improve chemical transport models (CTMs). The inorganic portion of aerosol, which includes both volatile (e.g., nitrate, ammonium) and non-volatile (e.g., calcium, sodium) species, controls many of these impacts through the regulation of charge balance, aerosol pH, and aerosol liquid water concentration (Guo et al., 2015, 2018; Hennigan et al., 2015; Nguyen et al., 2016; Pye et al., 2020). Further, the inorganic portion of aerosol is an important fraction of the aerosol budget, both in polluted cities (e.g., Jimenez et al., 2009; Song et al., 2018), and remote regions (e.g., Hodzic et al., 2020), and the chemistry controlling the inorganic portion of the aerosol is still not well known (e.g., Liu et al., 2020).

There are numerous methods to quantify the inorganic aerosol composition and mass concentration, including by mass spectrometry (DeCarlo et al., 2006; Canagaratna et al., 2007; Pratt and Prather, 2010; Froyd et al., 2019), *on-line* ion chromatography (Talbot et al., 1997; Weber et al., 2001; Nie et al., 2010), and collection onto filters to be extracted and measured off-line by ion chromatography (Malm et al., 1994; Dibb et al., 2002, 2003; Coury and Dillner, 2009; Watson et al., 2009). Each method has different advantages and disadvantages (e.g., time resolution, sample preparation, range of species identified, cost, and personnel needs). These results, in turn, have been used to inform and improve the results of CTMs, influencing our understanding in processes such as the direct radiative effect (Wang et al., 2008b), transport of ammonia in deep convection (Ge et al., 2018), aerosol pH (Pye et al., 2020; Zakoura et al., 2020) and subsequent chemistry, and precursor emissions (Henze et al., 2009; Heald et al., 2012; Walker et al., 2012; Mezuman et al., 2016).

Filter measurements have been shown to be most prone to artifacts during sample collection, handling, storage of the filter, or extraction of the aerosol from the filter prior to analysis. These artifacts include evaporation of volatile compounds such as organics (Watson et al., 2009; Chow et al., 2010; Cheng and He, 2015) and ammonium nitrate (Hering and Cass, 1999; Chow et al., 2005; Nie et al., 2010; Liu et al., 2014, 2015; Heim et al., 2020), as well as chemical reactions of gas-phase species with the accumulated particles (e.g., Schauer et al., 2003; Dzepina et al., 2007). Further, early research indicated potential artifacts from gas-phase ammonia uptake onto acidic aerosol collected onto filters, leading to a positive bias for particulate ammonium (Klockow et al., 1979; Hayes et al., 1980; Koutrakis et al., 1988). This led to debates about whether aerosol in the lower stratosphere was sulfuric acid or ammonium sulfate (Hayes et al., 1980); however, after improved filter handling practices and *on-line* measurements (i.e., mass spectrometry), it has been generally well accepted that the sulfate in the stratosphere is mainly sulfuric acid (Murphy et al., 2014).

This artifact may impact aerosol collected in remote locations (e.g., the lower stratosphere, but also the free troposphere over the Pacific Ocean basin). Comparisons for a major cation, ammonium, in a similar location (middle of the Pacific Ocean) have shown very different results (Dibb et al., 2003; Paulot et al., 2015). This, in turn, affects the observed charge balance of anions (sulfate and nitrate) with ammonium, which can indicate different aerosol phase state (Colberg et al., 2003; Wang et al., 2008a) and aerosol pH (Pye et al., 2020), leading to potentially important chemical and physical differences between the real state of the particles and that concluded from the measurements. An example of the differences in observed charge balance of ammonium to sulfate for different studies of the same remote Pacific Ocean region is highlighted in Fig. 1. This difference leads to the inorganic portion of the aerosol potentially being solid (filters) and hence good ice-nucleating particles (Abbatt et al., 2006), versus it being liquid (*on-line* measurements), leading to important differences in the calculated radiative balance. It should be noted that other measurements (both filter and *on-line*) in a similar location from another study (bar at surface (Paulot et al., 2015)) are more in-line with the *on-line* observations. A large decrease in the ambient ammonia mixing ratio is required to change from ammonium sulfate-like aerosols to sulfuric acid-like aerosols between the years, contradictory to the increasing trends of ammonia globally (Warner et al., 2016, 2017; Weber et al., 2016; Liu et al., 2019; Tao and Murphy, 2019). Further, oceanic emissions of ammonia are not high enough to lead to full charge neutralization of sulfate, since these emissions are approximately an order of magnitude less than those of sulfate precursors (Faloona, 2009; Paulot et al., 2015). A debate about the acidity and potential impact of ammonia-uptake artifacts on acidic filters for remote locations has not occurred as it did for stratospheric observations.

Previous laboratory studies have suggested that exposure of acidic aerosol, both suspended in air in a flow tube or on a filter, to gas-phase ammonia will lead to formation of ammonium salts in short time ($\leq$ 10 s) (Klockow et al., 1979; Huntzicker et al., 1980); however, it has not been investigated if this time frame applies for acidic aerosol collected on filters handled in a typical indoor environment. Though human emissions of ammonia are variable and depend on various factors (e.g., temperature, clothing, etc.) (Li et al., 2020), the emissions of ammonia, specifically from perspiration but also from breath, can lead to high, accumulated mixing ratios of ammonia indoor (e.g., Ampollini et al., 2019; Finewax et al., 2020) and references therein), depending on the ventilation rate. The mixing ratios of ammonia can be factor of 2 to 2000 higher indoor versus outdoor. This higher mixing ratio of ammonia leads to similarly high mixing ratios used in prior studies to lead to partially to fully neutralize sulfuric acid (Klockow et al., 1979; Huntzicker et al., 1980; Daumer et al., 1992; Liggio et al., 2011).

Here, we investigate whether previously observed laboratory observations of ammonium uptake to acidic particulate lead to the large differences in ammonium, both in mass concentration and in ammonium-to-sulfate ratios or ammonium-to-anion ratios, between *in-situ* measurements and *off-line* filter measurement during five NASA and one NSF airborne campaigns that sampled air over remote continental and oceanic regions. An uptake model for gas-phase ammonia interacting with acidic PM on a filter along with constraints from observations of gas-phase ammonia in the cabin of the airplane are used to further probe the reason behind the differences between the *in-situ* and *off-line* measurements of ammonium. The results provide insight into how to interpret prior aircraft measurements and other filter based measurements where the filters were handled in environments (i.e., indoors), where rapid uptake of ammonia to acidic PM will occur.

## 2. Methods

### 2.1 Aircraft Campaigns

Five different NASA aircraft campaigns on-board the DC-8 research aircraft and one NSF aircraft campaign on-board the C-130 research aircraft are used in this study. As described below, though the campaigns were sampling ambient (outside) air in various locations around the world, the filters were handled and exposed to both aircraft cabin air and indoor temporary laboratory air, where between 20 and 40 people were operating instruments. The campaigns include the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) -A (April 2008) and -B (June – July 2008) campaigns (Jacob et al., 2010), the Studies of Emissions and Atmospheric Composition, Clouds, and Climate Coupling by Regional Surveys (SEAC$^4$RS, August – September 2013) campaign (Toon et al., 2016), the Wintertime INvestigation of Transport, Emissions, and Reactivity (WINTER, February – March 2015) (Schroder et al., 2018), and the Atmospheric Tomography (ATom) -1 (July – August 2016) and -2 (January – February 2017) campaigns (Hodzic et al., 2020). ARCTAS-A was based in Fairbanks, Alaska, Thule, Greenland, and Iqaluit, Nunavut, and sampled the Arctic Ocean and Arctic regions of Alaska, Canada, and Greenland; while, ARCTAS-B was based in Cold Lake, Alberta, Canada, and sampled the boreal Canadian forest, including wildfire smoke. SEAC$^4$RS was based in Houston, Texas, and sampled biomass burning from western forest fires and agricultural burns along the Mississippi River and the Southern United States, isoprene chemistry over Southern United States and midwestern deciduous forests, and deep convection associated with isolated thunderstorms, the North American Monsoon, and tropical depressions. Finally, ATom-1 and -2 sampled the remote atmosphere over the Arctic, Pacific, Southern, and Atlantic Oceans during the Northern (Southern) Hemispheric summer (winter) and winter (summer).

For ARCTAS-A, -B, and SEAC$^4$RS, the general sampling scheme was regional, sampling large regions at level flight tracks. ATom-1 and -2, being global in nature, only sampled at level legs for short durations (5 – 15 min) at low (~300 m) and high (10 – 12 km) altitude, and did not measure at level altitudes between the low and high altitude. Due to the sampling time of the filters (see Sect. 2.2.2), the entirety of the ascent and descent time was needed for one filter sample. Therefore, all data during the ascents and descents have not been considered in this study to minimize any issues due to the mixing of aerosols of different compositions and acidities.

## 2.2 Aerosol Measurements

### 2.2.1 Aerosol Mass Spectrometer

An Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer, flown by the University of Colorado-Boulder (CU for short), was flown during the five campaigns used here. The general features of the AMS have been described in prior studies (DeCarlo et al., 2006; Canagaratna et al., 2007), and the specifics of the CU AMS for each campaign has been described elsewhere (Cubison et al., 2011; Liu et al., 2017; Nault et al., 2018; Schroder et al., 2018; Guo et al., 2020; Hodzic et al., 2020). In brief, the AMS measured the mass concentration of non-refractory species in $PM_1$ (PM with an aerodynamic diameter less than 1 μm, see Guo et al. (2020) for details). Ambient air was sampled by drawing air through an NCAR High-Performance Instrumental Platform for Environmental Modular Inlet (HIMIL; Stith et al. (2009)) at a constant standard flow rate of 9 L min$^{-1}$ (T = 273.15 K and P = 1013 hPa). The best estimated upper size cut-off for the HIMIL inlet is ~1 μm diameter (geometric, David Rogers, pers. comm. 2011). This diameter is larger than the size cut-off than that of the AMS inlet (~0.5-0.7 µm diameter, geometric, depending on the composition), with no losses in the tubing between the HIMIL and AMS inlet expected (see Guo et al. (2020) for more details). Multiple comparisons with instruments sampling from an isokinetic inlet $PM_4$ inlet (Brock et al., 2019; Guo et al., 2020) indicate that no significant sampling biases were incurred over the size range of the AMS. No active drying of the sampling flow was used to minimize artifacts for semi-volatile species, but the temperature differential between ambient and cabin typically ensured the relative humidity (RH) inside the sampling line less than 40% (e.g., Nault et al., 2018). An exception to this was during ATom-1 and -2, where the cabin temperature, along with the high RH in tropics, led to higher RH in the sample lines in a few instances in the boundary layer, which was accounted for in the final mass concentrations (Guo et al., 2020). To minimize any potential losses of volatile aerosol components, the residence time between the inlet and AMS was less than 1 s (Nault et al., 2018; Schroder et al., 2018; Guo et al., 2020). Prior studies (Guo et al., 2016; Shingler et al., 2016) have shown minimal loss of semivolatile components for this residence time.

The air sample was introduced into the AMS via an aerodynamic focusing lens (Zhang et al., 2002, 2004), which was operated at 2.00 hPa (1.50 Torr), via a pressure-controlled inlet, which was operated at various pressures (94-325 Torr) (Bahreini et al., 2008), depending on the ceiling of the campaign and lens transmission calibrations (Hu et al., 2017b; Nault et al., 2018). The aerosol, once focused, was introduced into a detection chamber after three differential pumping stages. The aerosol impacted on an inverted cone porous tungsten “standard” vaporizer under high vacuum, which was held at ~600°C. Upon impaction, the non-refractory portion of the aerosol (organic, ammonium, nitrate, sulfate, and chloride) were flash-vaporized, and the vapors were ionized by 70 eV electron ionization. The ions were then extracted and analyzed with a H-TOF time-of-flight mass spectrometer (Tofwerk AG). The AMS was operated in the "V-mode" ion path (DeCarlo et al., 2006), with spectral resolution ($m/\Delta m$) of 2500 at $m/z$ 44 and 2800 at $m/z$ 184. The collection efficiency (CE) for AMS was estimated with the parameterization of Middlebrook et al. (2012), which has been shown to perform well for ambient aerosols (Hu et al., 2017a, 2020). The AMS nominally samples aerosol with vacuum aerodynamic diameter between 40 nm and 1400 nm, which was calibrated for in SEAC$^4$RS, ATom-1, and -2 (Liu et al., 2017; Guo et al., 2020). Mass and/or volumen closure has been investigated between the AMS and other measurements for all campaigns discussed here (Cubison et al., 2011; Aknan, 2015; Liu et al., 2017; Nault et al., 2018; Schroder et al., 2018; Guo et al., 2020). The closure was complete for the size range of the AMS and did not show any dependence with altitude (Guo et al., 2020). Software packages Squirrel and PIKA under Igor Pro 7 (WaveMetrics, Lake Oswego, OR) (DeCarlo et al., 2006; Sueper, 2018) were used to analyze all AMS data.

A cryogenic pump, to reduce background of ammonium and organics (Nault et al., 2018; Schroder et al., 2018), was flown on the AMS for SEAC$^4$RS, ATom-1, and -2; but not for ARCTAS-A and -B. The cryogenic pump lowers the temperature of a copper cylinder surrounding the vaporizer to ~90 K. This freezes out the background gases and ensures low detection limits from the beginning of the flight, which is critical since aircraft instruments can typically not be pumped continuously and hence suffer from high backgrounds at switch-on. The

2σ accuracy for the AMS for inorganic aerosol is estimated to be 35% (Bahreini et al., 2009; Guo et al., 2020).

**2.2.2 Aerosol Filters**

Fast collection of aerosol particles onto filters during airborne sampling, via the University of New Hampshire Soluble Acidic Gases and Aerosol (SAGA) technique, has been described elsewhere (Dibb et al., 2002, 2003), and was flown during the five campaigns investigated here. Briefly, air is sampled into the airplane via a curved leading edge nozzle (Dibb et al., 2002). The inlet is operated isokinetically during flight, and typically has a 50% collection efficiency for aerosol with an aerodynamic diameter of 4.1 µm (Dibb et al., 2002; McNaughton et al., 2007), with some altitude dependence (Guo et al., 2020). The lower size cut-offs for SAGA and AMS are similar (Guo et al., 2020). As discussed by Guo et al. (2020; their Fig. 8) the difference in mass sampled at the smaller sizes between SAGA and AMS is generally negligible at all altitudes.

Aerosol was collected onto Millipore Fluoropore Teflon filters (90 mm diameter with 1 µm pore size). Collection time was dependent on altitude and estimated mass concentration, but generally 2 to 3 $sm^3$ (where $sm^3$ is standard $m^{-3}$ at temperature = 273 K and pressure = 1013 hPa) volume of air is collected to ensure detectable masses of species (Dibb et al., 2002). The aerosol inlet flow is close to 400 slpm in the marine boundary layer and approximately 150 slpm at maximum altitude. Further, 2 blank filters are collected each flight. The filters were contained in a Delrin holder during collection. After collection, the filters were transferred to a particle free polyethylene "clean room" bag, which was filled with zero air, sealed, and stored over dry ice.

No acid scrubbers were inserted into the bags to prevent any artifact from offgassing of ammonia. The samples from the filters were then extracted during non-flight days with 20 mL ultrapure water and preserved with 100 µL chloroform (see Sect. S2). The preserved samples were sent to the University of New Hampshire, to be analyzed by ion chromatography. The estimated limit of detection for both sulfate and ammonium is 0.01 µg $sm^{-3}$ for all missions evaluated here (Dibb et al., 1999).

### 2.2.3 Other Aerosol Measurements

The NOAA Particle Analysis by Laser Mass Spectrometer (herein PALMS) was flown during ATom-1 and -2. Details of the PALMS instrument configured for ATom-1 and -2 are described in Froyd et al. (2019). Briefly, PALMS measures the chemical composition of single aerosol particles via laser-ablation/ionization (Murphy and Thomson, 1995; Thomson et al., 2000), where the ions are extracted and detected by a time of flight mass spectrometer. The instrument measures particles between 100 nm and 4.8 µm (geometric diameter) (Froyd et al., 2019). The measurement of PALMS used in this study is the "sulfate acidity indicator" (Froyd et al., 2009). These authors reported that in the negative ion mode, there is a prominent peak at *m*/*z* 97, corresponding to $HSO_4^-$, and another peak at *m*/*z* 195, corresponding to the cluster $HSO_4^-(H_2SO_4)$. The first peak was independent of acidity; whereas, the second peak was dependent on acidity. Froyd et al. (2009) calibrated the PALMS ratio of $HSO_4^-(H_2SO_4)/(HSO_4^-+HSO_4^-(H_2SO_4))$ to Particle-into-Liquid Sampler (PILS) measurements to achieve an estimate of ammonium balance.

Besides the chemical composition, the particle number and volume distributions are used here. For SEAC$^4$RS, the measurements have been described elsewhere (e.g., Liu et al., 2016). The laser aerosol spectrometer (from TSI), which measured aerosol from geometric diameter 100 nm to 6.3 µm, is used here for volume distribution. For the ATom missions, the measurements have been described elsewhere (Kupc et al., 2018; Williamson et al., 2018; Brock et al., 2019). Briefly, the dry particle size distribution, from geometric diameter of 2.7 nm to 4.8 µm, were measured by a series of optical particle spectrometers, including the Nucleation Model Aerosol Size Spectrometer (3 nm to 60 nm, custom built (Williamson et al., 2018)), an Ultra-High Sensitivity Aerosol Spectrometer (60 nm to 1 µm) from Droplet Measurement Technologies (Kupc et al., 2018), and Laser Aerosol Spectrometer (120 nm to 4.8 µm) from TSI). These measurements have been split in nucleation mode (3 to 12 nm), Aitken mode (12 to 60 nm), accumulation mode (60 to 500 nm) and coarse mode (500 nm to 4.8 µm).

## 2.3 Gas-Phase and Other Measurements

### 2.3.1 Ammonia Measurements

Gas-phase ammonia was measured inside the cabin of the NASA DC-8 during the FIREX-AQ campaign (Warneke et al., 2018), a subsequent DC-8 campaign which shared many instrument installations and a similar level of aircraft personnel with the campaigns analyzed here. The location of the instrument and where it sampled cabin ammonia (in relation to where the SAGA filters are located) is shown in Fig. S1. Ammonia was measured by a Picarro G2103 Gas Concentration Analyzer (von Bobrutzki et al., 2010; Sun et al., 2015; Kamp et al., 2019). The instrument is a continuous, cavity ring-down spectrometer. Cabin air is brought into a cavity at low pressure (18.7 kPa, 140 Torr), where laser light is pulsed into the cavity. The light is reflected by mirrors in the cavity, providing an effective path length of kilometers. A portion of the light penetrates the mirrors, reaching the detectors, where the intensity of the light is measured to determine the mixing ratio of ammonia from the time decay of the light intensity via Beer-Lambert Law. The instrument measures the absorption of infrared light from 6548.5 to 6549.2 $cm^{-1}$ (Martin et al., 2016). Absorption of gas-phase water is also measured and corrected for. This water vapor measurement is also used to calculate RH inside the cabin of the DC-8 (Filges et al., 2018). Data was logged at 1 Hz.

**2.3.2 Carbon Dioxide and Temperature Measurements**

Carbon dioxide inside the cabin of the NASA DC-8 during FIREX-AQ was measured by a HOBO MX1102 Carbon Dioxide Data Logger (HOBO by Onset). It is a self-calibrating carbon dioxide sensor with a range of 0 to 5,000 ppm carbon dioxide and an accuracy of ±50 ppm. A non-dispersive infrared sensor is used to measure carbon dioxide. Data was acquired once every 10 s to once every 2 min. Besides carbon dioxide, RH and temperature are also recorded by the instrument. Prior to each flight, the instrument was turned on and measured ambient carbon dioxide, outside the cabin of the DC-8, to ensure the accuracy of the instrument compared to ambient carbon dioxide measurements.

Ambient carbon dioxide during FIREX-AQ was measured by an updated version of the instrument known as Atmospheric Vertical Observations of $CO_2$ in the Earth's Troposphere (AVOCET) (Vay et al., 2003, 2011). The updated instrument used a modified LI-COR model 7000 non-dispersive infrared spectrometer and measured carbon dioxide at 5 Hz.

Temperature in the cabin was measured by a thermocouple (SEAC$^4$RS) or thermistor (ATom-1 and 2) located in the AMS rack or a Vaisala probe located at the front of the airplane (ARCTAS-A, -B, and SEAC$^4$RS).

## 2.4 Theoretical Ammonia Flux Model

To investigate the possibility that the ammonia mixing ratio in the cabin of the DC-8 is high enough to be taken up by acidic PM on a filter during the short time the filter is exposed to cabin air prior to final storage, a theoretical uptake model was constructed to estimate the time scale for ammonia to interact with all the acidic particles (Seinfeld. and Pandis, 2006). The equations used for the model can be found in the Supplemental Information (Sect. S3). The model was initialized with a range of ammonia mixing ratios (1 to 200 ppb) and a range of PM diameters (10 to 1000 nm). The calculations were conducted at 298 K, which is within ±10 K of typical temperatures inside the cabin of the NASA DC-8 during the five campaigns (Fig. S2). An accommodation coefficient of 1 for ammonia onto acidic PM was assumed (Hanson and Kosciuch, 2003), with a density of 1.8 g cm$^{-3}$ for sulfuric acid (Rumble, 2019). For the mass transfer calculations, the transition regime (between the free molecular and continuum regimes) equations were used, using the Fuchs and Sutugin parameterization (Fuchs and Sutugin, 1971). The model was used to estimate the ammonia molecular flux to acidic PM on the filter (Eq. S3). Finally, the molecular flux was used to estimate the time it would take all the particles to be partially neutralized by ammonia in the cabin (Eq. S4), though this may be a lower limit (Robbins and Cadle, 1958; Daumer et al., 1992).

## 3. Results and Discussion

### 3.1 Comparison of On-Line and Off-Line Ion Balances across the Tropospheric Column

SAGA and AMS co-sampled aerosols during multiple aircraft campaigns. Nitrate quickly evaporates from aerosols as the aerosols are transported away from source regions and is typically small in the global troposphere (DeCarlo et al., 2008; Hennigan et al., 2008; Hodzic et al., 2020). Thus, in Fig. 2 the mass concentrations for the two most important submicron contributors to ammonium balance, ammonium and sulfate, are compared from the aircraft campaigns. The campaigns generally sampled remote air, either continental or oceanic, except for biomass burning sampled during ARCTAS-B and SEAC$^4$RS and downwind of urban areas during WINTER. The measurements, for mass concentrations greater than 0.1 μg $sm^{-3}$, are generally within the combined uncertainties of the two instruments. Sulfate generally remains on the one-to-one line, even at low mass concentrations. However, ammonium shows a large divergence between the two measurements for mass concentrations less than 0.1 μg $sm^{-3}$ during all six aircraft campaigns. As shown in Fig. 2, the divergence in ammonium occurs well above the limit-of-detection for both instruments, namely ~4 ng $sm^{-3}$ for AMS for a 5-minute average (DeCarlo et al., 2006; Guo et al., 2020) and 10 ng $sm^{-3}$ for SAGA (Dibb et al., 1999), for both ammonium and sulfate.

This divergence in ammonium mass concentration is thus reflected in the ammonium balance, defined as the ratio of ammonium to sulfate plus nitrate, in moles (Fig. 3). For all campaigns, the two measurements show differences in ammonium balance, especially at higher altitudes, where the aerosols is distant from ammonia emissions (Dentener and Crutzen, 1994; Paulot et al., 2015), but sulfate production can continue due to vertical transport of precursors such as $SO_2$. On average, the SAGA measurements indicate ammonium balance rarely below 0.5 throughout the troposphere; whereas, the AMS measurements indicate that ammonium balance generally drops to below 0.2 for pressures less than 400 hPa. Fig. 2 and Fig. 3 indicate either differences in the ammonium balance due to differences in aerosols population sampled, as SAGA measures larger aerosols diameters than AMS (Guo et al., 2020), or potential artifacts with one of the measurements.

Both the AMS and the filters sample most of the submicron aerosols (see Guo et al. (2020) for details), but the filters also sample supermicron particles that the AMS does not. Therefore it is possible in principle that the difference could be due to ammonium present in supermicron particles. As discussed in Guo et al. (2020), nearly 100% of the measured volume occurs for aerosols < 1 μm above the marine boundary layer, where the largest difference in ammonium balance between the filters and AMS occurs (Fig. 3). Further, ammonium has been observed to be a small fraction of the supermicron mass (Kline et al., 2004; Cozic et al., 2008; Pratt and Prather, 2010), except for instances of continental fog (Yao and Zhang, 2012) and Asian dust events (Heim et al., 2020). An upper estimate of supermicron ammonium can be calculated using results from prior studies (Kline et al., 2004; Cozic et al., 2008). In these prior studies, ~90% of the ammonium was submicron. With the average ammonium observed during ATom-1 and -2 (~10 to 50 ng $sm^{-3}$) (Hodzic et al., 2020), that would suggest an upper limit of ~1 to 5 ng $sm^{-3}$ ammonium in the supermicron aerosols. This upper estimate does not explain the differences between AMS and filters during ATom-1 and -2 (Fig. S3), as the percent difference increases with decreasing estimated supermicron ammonium volume. As the largest differences between the AMS and filters occur well above the boundary layer (Fig. 3), away from continental ammonia sources (Dentener and Crutzen, 1994) and Asian dust events, we conclude that the sampling of supermicron aerosols by filters is not leading to the observed differences in ammonium.

The only useful comparison, other than SAGA versus AMS, is with PALMS during ATom. Prior studies by PALMS have shown aerosols observed for pressure < 400 hPa to be acidic, depending on potential recent influence of boundary layer air via convection (Froyd et al., 2009; Liao et al., 2015), similar to observations by other single particle mass spectrometers (Pratt and Prather, 2010). Though not reaching similarly low $NH_4/(2\times SO_4)$ values as the AMS, the PALMS acidity marker shows much lower values than were determined by the aerosols collected on the filters (Fig. S4). Different reasons for PALMS not achieving as low values as AMS may include differences in aerosols sizes sampled by PALMS versus AMS (Guo et al., 2020), and the sensitivity of the acidity marker to laser power (Liao et al., 2015). Thus, two different *on-line* measurements indicate that the ammonium balance is lower than the aerosols collected on filters, suggesting potentially more acidic aerosols.

Differences in ammonium balance between AMS and SAGA are detectable for sulfate mass concentrations $\leq 1$ μg $sm^{-3}$ (Fig. 4) for all six aircraft campaigns. As the sulfate mass concentration decreases, the relative differences in ammonium, and thus ammonium balance, increase. The large majority of the troposphere contains sulfate mass concentrations in which the differences in ammonium are observed, highlighting the importance of this problem (Fig. 4a). Thus, except for more polluted conditions (> 1 μg $sm^{-3}$ sulfate), which mainly occurs in continental (Jimenez et al., 2009; Kim et al., 2015; Malm et al., 2017) and urban regions (Jimenez et al., 2009; Hu et al., 2016; Kim et al., 2018; Nault et al., 2018), this bias between filters and *on-line* measurements is critically important, especially since airborne measurements are often the only meaningful observational constraints for remote regions. Thus, this analysis suggests that for SAGA filters , a more meaningful ammonium limit-of-detection would be equivalent to 1 μg $sm^{-3}$ sulfate, which would be ~0.2 μg $sm^{-3}$ ammonium. This also provides the framework to define limit-of-detection for other filter-based measurements not associated with ion chromatography.

### 3.2 Ammonia Levels on the NASA DC-8 Cabins

Prior studies have suggested that various sources of ammonia could impact acidic filter measurements (Klockow et al., 1979; Hayes et al., 1980; Koutrakis et al., 1988). Some of these studies found that the materials of the containers where the filters are stored, unless thoroughly cleaned and not stored around humans, are a source of ammonia gas that reacts with the sulfuric acid on the filters to become ammonium, leading to ammonium bisulfate or ammonium sulfate (Hayes et al., 1980). Further, handling of acidic filters in rooms with people or acidic aerosol in the presence of human breath can also lead to near to complete neutralization of acidic aerosol (Larson et al., 1977; Hayes et al., 1980; Clark et al., 1995). Finally, various studies have suggested that the SAGA filters specifically may be impacted by various ammonia sources prior to sampling with the ion chromatography (Dibb et al., 1999, 2000; Fisher et al., 2011).

During SAGA sampling, the filters with collected aerosol are moved from the sample collector to a polyethylene bag that is filled with clean air. During this step, the filter is exposed to the cabin air of the DC-8 for ~10 s. As humans are a source of ammonia (Larson et al., 1977; Clark et al., 1995; Sutton et al., 2000; Finewax et al., 2020; Li et al., 2020), this source sustains significant ammonia concentrations in indoor environments, which could potentially bias the filter measurements. *On-line* measurements would not be subject to this effect since the sampled air is not exposed to cabin air before measurement. While inlet lines for *on-line* instruments could in theory lead to some memory effects, there is no evidence of such effects in the data (e.g., the response going from a large, neutralized plume into the acidic FT is nearly instantaneous (Schroder et al., 2018)).

During a recent 2019 NASA DC-8 aircraft campaign, FIREX-AQ, ammonia was measured on-board the DC-8 during several research flights. An example time series of cabin ammonia, temperature, and RH is shown in Fig. 5. Prior to take-off, as scientists were slowly boarding the airplane, the ammonia mixing ratio was low (< 20 ppbv) and similar to ambient levels of ammonia outside the aircraft. As scientists started boarding before take-off, the ammonia mixing ratio increased. Upon doors closing, the mixing ratio leveled off at ~40 ppbv. After take-off, the mixing ratio remained ~40 ppbv, though there were changes related to changes in cabin temperature and humidity, which would affect emission rates and also adsorption of ammonia onto cabin surfaces (Sutton et al., 2000; Finewax et al., 2020; Li et al., 2020) and movement of scientists throughout the cabin, which would affect emission rates and their location.

The average (±1σ spread of the observations) and median ammonia in the cabin of the DC-8 during FIREX-AQ was 45.4±19.9 and 41.9 ppbv (Fig. 6). There was a large positive tail in ammonia mixing ratio, related to high temperatures (Fig. S5), which causes the scientists to perspire more and release more ammonia (Sutton et al., 2000; Finewax et al., 2020; Li et al., 2020). Compared to outdoor ammonia mixing ratios, ranging from urban to remote locations, the ammonia in the cabin of the DC-8 is higher by a factor of 2 to 2000 (Fig. 6). On the other hand, the ammonia measured in the cabin of the DC-8 is similar but towards the lower end of the mixing ratios measured during various indoor studies (Table S1 for compilation of references).

The ammonia mixing ratios observed in the cabin were verified by investigating the cabin air exchange rates (see SI Sect. S3). Using carbon dioxide measurements, the exchange rate in the cabin was calculated to be 9.9 $hr^{-1}$ (Fig. S6), which is similar to literature values for the cabin exchange rate of other passenger airliners (Hunt and Space, 1994; Hocking, 1998; Brundrett, 2001; National Research Council, 2002). This value is a factor of 2 to 5 higher than typical exchange rates for commercial buildings (Hunt and Space, 1994; Pagonis et al., 2019), which would suggest lower mixing ratios than observed in other indoor environments. Using this exchange rate, and the literature total ammonia emission rates from humans (1940 $\mu g\ hr^{-1}\ person^{-1}$ (Sutton et al., 2000)) and the average of ambient ammonia mixing ratios as an outdoor background onto which the human emissions in the cabin are added (~4.4 ppbv, Fig. 6), the ammonia mixing ratio in the cabin of the DC-8 was estimated to be 43.4 ppbv, which is within the uncertainty of the average ammonia (45.4±19.9 ppbv) inside the cabin of the DC-8. Thus, the observed ammonia mixing ratios in the cabin of the DC-8 are consistent with the cabin air exchange rates and literature human ammonia emissions. These mixing ratios are approximately a factor of nine higher than in a typical laboratory environment (Fig. S7), as there are fewer people (1 to 4 versus 20 to 40), making the cabin of the DC-8 an extreme laboratory environment for handling acidic filters. As shown in Fig. 6, ammonia mixing ratios in indoor environments are high enough to change the thermodynamics of inorganic aerosol, leading to higher ammonium balances (Weber et al., 2016). Thus, similar to the conclusions of other studies, the cabin of the DC-8 is an important source of ammonia that could lead to biases with acidic aerosols collected on filters.

During FIREX-AQ, the DC-8 frequently sampled air impacted by biomass burning, which is an important source of ammonia (Sutton et al., 2013) and could potentially increase the background ammonia being brought into and mixing with the cabin air being sampled by the Picarro. Splitting the cabin ammonia ratios between sampling air impacted by biomass burning versus nominally background air, the normalized PDF did not shift to higher ammonia mixing ratios (Fig. S7). Further, the averages of the observed cabin ammonia was statistically similar, at the 95% confidence interval, between the DC-8 sampling biomass burning and nominally background air (48.1±13.4 versus 44.1±14.4 ppbv for biomass burning and background air, respectively). Finally, the majority of the time the cabin air was sampled by the Picarro for cabin ammonia, the DC-8 was sampling agricultural fires in Southeast US, which are shorter in duration (seconds) versus the large wildfires in Western US (minutes to hours). This is reflected in the low average ambient value for ammonia, as measured by a proton transfer reaction mass spectrometer (Müller et al., 2014), when the DC-8 was sampling biomass burning-influenced air observed during this time (~10 ppbv) and very low average value for non-biomass burning-influenced air (~08.8 ppbv) (Fig. S7). Thus, ammonia from biomass burning would at most be a small impact on the ammonia measured in the cabin of the DC-8, further indicating the ammonia in the cabin was mainly from human emissions.

### 3.3 Can Uptake of Cabin Ammonia Explain the Higher Ammonium Concentrations on Filters?

As shown in Fig. 6, the cabin of the DC-8 is an important source of ammonia from the breathing and perspiring of scientists. Prior studies (Klockow et al., 1979; Huntzicker et al., 1980; Daumer et al., 1992; Liggio et al., 2011) have shown in laboratory settings that 10 s is fast enough to partially to fully neutralize sulfuric acid. Thus, here we investigate whether the time of the filter handling of 10 s will lead to partial to full neutralization of sulfuric acid from cabin ammonia, or whether this time is fast enough to limit exposure of the acidic filter to cabin ammonia. Huntzicker et al. (1980) showed that for typical aerosol modal distributions (Fig. 7) and cabin RH (Fig. S9), an initial pure sulfuric acid aerosol, suspended in a flow reactor, reaches equal molar amounts of ammonium and sulfate (i.e., ammonium bisulfate) when exposed to 70 ppb ammonia in 10 s. This indicates the plausibility that acidic aerosol filters, which typically have lower sulfate mass concentrations than Huntzicker et al. (1980) (~2 μg versus ~55 μg sulfate equivalent on filters), would interact with cabin ammonia to form at least ammonium bisulfate. Further, other studies found that in less than 10 s, sulfuric acid aerosol, suspended in a flow reactor, at RH ≤ 45%, will completely react with gas-phase ammonia to form ammonium sulfate (Robbins and Cadle, 1958; Daumer et al., 1992). The latter study used ammonia mixing ratios similar to the amount observed in the cabin of the DC-8 (~30 ppbv); whereas, the former study used excess ammonia (~9 ppmv). Some studies have suggested that the bags used to store the filters may be a source of ammonia (e.g., Hayes et al., 1980); however, calculations indicate the bags would be a small source of ammonia (see Sect. S4).

First, the time of diffusion of ammonia gas from the surface to the interior of the filter was investigated, as there is a potential for the PM to be embedded deep into the filter. Eq. 1 (Seinfeld. and Pandis, 2006):

$$\tau_{diffusion} = \frac{d_t^2}{8D_g} \quad \text{Eq. 1}$$

where $d_t^2$ is the depth of the Teflon (~0.015 cm) and $D_g$ is the diffusion coefficient of ammonia in air (0.228 $cm^2$ $s^{-1}$) (Spiller, 1989). Therefore, the estimated timescale for ammonia to diffuse through the depth of the Teflon filter is ~$1\times10^{-4}$ s, meaning that the surface of PM will always be in contact with cabin-level mixing ratios of ammonia. Even though the filters have a porous membrane, for molecular diffusion, the membrane only increases the pathway that the ammonia molecules have to travel slightly; thus, not changing the estimated time. Second, as the particles are liquid (Wilson, 1921), the diffusion will be similar as through water. A typical value for diffusivity in water is ~$1\times10^{-5}$ $cm^2$ $s^{-1}$ (Seinfeld. and Pandis, 2006). For the size ranges observed (Fig. 7, ~40 - 700 nm), this corresponds to a timescale of $1.6\times10^{-7}$ to $5.0\times10^{-5}$ s. Thus, the diffusion through the filter and through the PM is nearly instantaneous for ammonia.

A theoretical uptake model for ammonia to acidic PM on filters was run for a range of ammonia mixing ratios and PM diameters (Fig. 7). As shown in Fig. 7, only at the lowest ammonia mixing ratios (< 10 ppbv), the flux of ammonia to acidic PM is slower (> 20 s) than the typical filter handling time (~10 s) for typical aerosol diameters in the remote atmosphere. For the conditions of the DC-8, similar to other indoor environments (> 20 ppbv ammonia, Fig. 6), and ambient aerosol diameters in the accumulation mode that contains most ambient sulfate (Fig. 7), the amount of time needed for cabin ammonia to interact with acidic PM on filters to form ammonium bisulfate is ≤ 10 s, similar to the results of Huntzicker et al. (1980). Also, studies show that the kinetic limitation to form ammonium sulfate ($(NH_4)_2SO_4$) versus ammonium bisulfate ($NH_4HSO_4$) is relatively low and can occur within the 10 s time frame (Robbins and Cadle, 1958; Daumer et al., 1992). A laboratory setting with ~5 ppbv $NH_3$ would result in the filters needing to be exposed to laboratory air for at least 40 s to form ammonium bisulfate (Fig. S8) versus the 3 to 10 s for conditions in the cabin of the DC-8 (Fig. 7), further exemplifying the challenging conditions of the DC-8 cabin for filter sampling.

The prior analysis made the assumption that the PM maintained a spherical shape upon impacting the Teflon filter. More viscous (i.e., solid) PM is more likely to maintain a spherical shape on filters whereas less viscous (i.e., liquid) PM will spread and become more similar to cylindrical shape (e.g., Slade et al., 2019). As more acidic aerosol is more likely to be liquid (e.g., Murray and Bertram, 2008), an exploration of cylindrical shape was conducted. Depending on the assumed height of the cylindrical shape, the timescale for a molecule of ammonia to interact with a molecule of sulfuric acid decreases from ~5 s (for maximum ammonia and aerosol volume) to ~4 s (assuming height of cylinder equals radius of sphere) to less than 1 s as height decreases from 25 nm or less. The aerosol deforming and spreading upon impacting the filters increases the particle surface area, and decreases the amount of time for cabin ammonium to interact with the acidic PM. Thus, less viscous aerosol has more rapid uptake and interaction with ammonia due to the higher surface area.

A potential limitation to the model is the accommodation coefficient of ammonia to acidic PM, as there are conflicting reports on its value (Hanson and Kosciuch, 2004; Worsnop et al., 2004). However, as shown in Worsnop et al. (2004), once the sulfuric acid weight percentage is 50% or greater, the different studies converge to an accommodation coefficient of ~1. Various studies indicate that the RH in the cabin of jet airplanes is low due to how air is brought into the airplane, typically $< 20\%$ (Hunt and Space, 1994; Brundrett, 2001; National Research Council, 2002). Even though the ambient RH may be higher than the RH in the cabin of the DC-8, the water equilibration is rapid (< 1 s) for the temperature of the cabin of the DC-8, even for very viscous aerosol (Shiraiwa et al., 2011; Price et al., 2015; Ma et al., 2019), meaning the PM on the filter would rapidly reach equilibrium with the cabin RH upon exposure. This would result in a ≥ 60% sulfuric acid weight percentage (Wilson, 1921) for the typical RH ranges in the cabin of typical airlines. However, various measurements in the DC-8 cabin indicate the RH is ≤ 40% (Fig. S9), leading to sulfuric acid weight percentage of 50% or greater (Wilson, 1921). Therefore, the accommodation coefficient of ~1 is well-constrained by the literature. Thus, the handling of the filters between the sampling inlet to the polyethylene bag exposes the acidic PM to enough gas-phase ammonia towards forming ammonium bisulfate or ammonium sulfate, biasing high ammonium from the filters. This explains the differences seen in Fig. 1 – Fig. 4.

Another potential limitation is that the PM on the filters could form a layer, as multiple particles pile up on top of each other, slowing the diffusion of ammonia to be taken up by acidic PM. The filters have a one-sided surface area of $6.4\times10^{-3}$ $m^2$, while an individual particle at the mode of the volume distribution (Fig. 7) has a projected surface area of ~$7.1\times10^{-14}$ $m^2$. Thus, ~$9.0\times10^{10}$ particles would need to be collected to form a single layer of PM on the filter. The number of molecules in a single particle of the mode size is ~$1.4\times10^{8}$ molecules (Eq. S2). Therefore, ~$1.3\times10^{19}$ molecules need to be collected onto the filters in order to form a monolayer of PM, which is equivalent to ~$2.2\times10^{3}$ μg total aerosol collected or ~700 μg $sm^{-3}$ aerosol concentration. As the mass concentration in ATom was typically ~1 μg $sm^{-3}$ (Hodzic et al., 2020), and total aerosol concentrations that high is rarely seen except for extreme events (such as the thickest fresh wildfire plumes), it is very unlikely that more particle layering would delay the diffusion of ammonia to acidic PM.

Various sensitivity analyses of the uptake of ammonia to sulfuric acid were conducted. First, there is minimal impact of cabin temperature on the results. Though there was a 25 K range in cabin temperature (Fig. S2), the impact on the molecular speed of ammonia in the model (Eq. S1) leads to a ±2% change in molecular speed, resulting in small changes in the time. Further, only large changes in the accommodation coefficient with temperature occurs for sulfuric acid weight percentages < 40% (Swartz et al., 1999), which is smaller than the weight percentage expected for the filters in the cabin of the DC-8. For the temperature range of the cabin of the DC-8 (Fig. S2), the coefficient changes by less than 10%, which leads to a total maximum change in Fig. 7 of ±12%. The largest impact on the results in Fig. 7 is changing the accommodation coefficient. Reducing the accommodation coefficient by a factor of 10, though not representative of the DC-8 cabin conditions, would mean the acidic PM would need to be exposed to ammonia for ≥ 1 minute (Fig. S10). It is expected that the lower accommodation coefficient will occur for conditions with higher RH (>80%), suggesting typical laboratory conditions (along with the lower ammonia mixing ratios) or ambient conditions may experience lower ammonia uptake to acidic PM. Finally, organic coatings may impact the accommodation coefficient of ammonia to sulfuric acid; however, the amount of reduction on the accommodation coefficient has varied among studies (e.g., Daumer et al., 1992; Liggio et al., 2011). Daumer et al. (1992) showed no impact; whereas, Liggio et al. (2011) found a similar impact to reducing the accommodation coefficient by a factor of 10 (Fig. S10). Thus, the results in Fig. 7 are in line with Daumer et al. (1992) while the results in Fig. S10 are in line with Liggio et al. (2011).

### 3.4 Impacts of Ammonia Uptake on Acidic Filters

As discussed throughout this study, uptake of cabin ammonia during the handling of acidic filters can lead to biases in ammonium mass concentrations. However, other potential sources of biases include the material used for sampling and storing the filter (Hayes et al., 1980), and the preparation of the filter in the field to be sampled by ion chromatography. As the preparation of the filters occurs indoors, as well, the filters will be exposed to similar ammonia mixing ratios to those shown in Fig. 6.

Further, filter collection of aerosols is a widely used technique outside of aircraft campaigns, including for regulatory purposes and long-term monitoring at various locations around the world. For many of these sites, ammonia denuders are used to minimize biases of ammonium on filters (e.g., Baltensperger et al., 2003). Data from remote, high altitude locations have indicated that the ammonium balance is less than one (Cozic et al., 2008; Sun et al., 2009; Freney et al., 2016; Zhou et al., 2019), similar to the observations from the AMS shown in Fig. 3. However, this is dependent on air mass origin and type (Cozic et al., 2008; Sun et al., 2009; Fröhlich et al., 2015). Thus, sampling of remote aerosols with filters does provide evidence of ammonium balances less than one due to a combination of procedures to minimize interaction of gas-phase ammonia with the acidic filters and the lower human presence (and potentially cooler temperatures at high, remote, mountaintop locations such as Jungfraujoch).

However, there are some long-term monitoring stations that do not use denuders or other practices to minimize the interaction of ammonia with acidic aerosols. For example, the Clean Air Status and Trends Network (CASTNET), which is located throughout the continental United States, measures ammonium, sulfate, and nitrate (Solomon et al., 2014). The CASTNET system uses an open-face system to collect aerosols on Teflon filters for approximately one week for each filter (Lavery et al., 2009). In comparison, the Chemical Speciation Monitoring Network (CSN), which also samples the continental United States and collects the aerosols on Nylon or Teflon filters, a denuder is used to scrub gas-phase ammonia to minimize interaction of ammonia with acidic aerosols on filters (Solomon et al., 2000, 2014). The comparison between these two long-term monitoring sites show very different trends of ammonium balance versus total inorganic mass concentration (Fig. S11). For CSN, the ammonium balance decreases with mass concentration whereas CASTNET remains nearly constant. This is similar to the comparison between SAGA and AMS in Fig. 4. This difference between the two sampling techniques may be due to the lack of denuder in CASTNET to remove gas-phase ammonia. The use of the denuders has led to CSN and other monitoring networks that use denuders to be more in-line with in-situ observations (Kim et al., 2015; Weber et al., 2016). Further, as shown in Fig. S8, exposure of an unprotected acidic filter for time greater than 1 day will lead to ammonia reacting with the acid to form ammonium bisulfate or ammonium sulfate, even at low ammonia mixing ratios. Other aspects that could impact this comparison, and are beyond the scope of this study (but has been discussed in other studies (Hering and Cass, 1999; Schauer et al., 2003; Chow et al., 2005, 2010; Dzepina et al., 2007; Watson et al., 2009; Nie et al., 2010; Liu et al., 2014, 2015; Cheng and He, 2015; Heim et al., 2020)), include the loss of volatile ammonium from the evaporation of ammonium nitrate or differences in the handling, shipping, and/or storage of the filters or extracted samples. Thus, without denuders, or handling of filters with more than one person present, will lead to similar differences between in-situ sampling versus filter collection of inorganic aerosols observed during various aircraft campaigns.

Further, the uptake of ammonia onto acidic aerosols will impact comparisons with chemical transport models (CTMs) and the understanding of various physical processes. For example, various CTMs predict different results for the mass concentration of ammonium in the upper troposphere (Wang et al., 2008a; Fisher et al., 2011; Ge et al., 2018), and selection of one measurement versus the other will lead to different degrees of agreement. For example, for filters that collect aerosols similar to those described here (no ammonia scrubber and/or exposed to human emissions of ammonia), values of ammonium < 0.2 μg $sm^{-3}$ should be used with caution or instead use *on-line* measurements of ammonium (specifically for SAGA measurements but a similar analysis should be conducted for other filter-based measurements). This different agreement impacts our understanding of important processes, such as the direct radiative impact of inorganic aerosol (Wang et al., 2008b) or deposition of inorganic gases and aerosols (Nenes et al., 2020a), as the gas-phase species have a faster deposition rate than the aerosol-phase. Finally, the measurement biases can impact the suggested regulations to improve air quality (Nenes et al., 2020b) and the calculated aerosol pH, as the pH is sensitive to the partitioning of ammonia between the aerosol- and gas-phase (e.g., Hennigan et al., 2015).

**Conclusions**

Collection of aerosols onto filters to measure aerosol mass concentration and composition is valuable for improving our understanding of the emissions and chemistry of inorganic aerosol, and longstanding, multi-decadal filter-based records of atmospheric composition are invaluable to analyze atmospheric change. However, as had been discussed in earlier studies, acidic aerosols collected on filters are susceptible to uptake of gas-phase ammonia, which interacts with the acidic aerosol to form an ammonium salt (e.g., ammonium bisulfate or ammonium sulfate). This artifact in filter measurements can bias our understanding on the chemical composition of the aerosol, which impacts numerous atmospheric processes.

We show that across six different aircraft campaigns, the aerosol collected on filters showed a substantially higher ammonium mass concentration and ammonium balance compared to AMS measurements. Further, another *on-line* measurement (PALMS) also shows lower ammonium-to-sulfate ratios than for the filters. These differences are not due to differences in the aerosol size ranges sampled by the PALMS and the filters. Instead, we show that the mixing ratio of gas-phase ammonia in the cabin of the DC-8 is high enough (mean ~45 ppbv), and similar to other indoor environments, to interact with acidic aerosol collected on filters in ≤ 10 s, to form ammonium salts. These results are consistent with prior studies investigating this interference. Thus, due to the interaction of ammonia in the cabin of research aircraft, we suggest that a more realistic limit-of-detection of ammonium for the SAGA filters is 200 ng $sm^{-3}$, versus the 10 ng $sm^{-3}$ typically cited based on the ion chromatography measurement. Finally, even though methods to reduce this bias have been implemented in several ground-based long-term filter measurements of inorganic aerosols, there are still some networks that collect inorganic aerosol without denuders to remove gas-phase ammonia, leading to similar discrepancies between ground networks as observed between filters and AMS on the various aircraft campaigns. Careful practice in both the aerosol collection and filtering handling is necessary to better understand the emissions, chemistry, and chemical and physical properties of inorganic aerosol.

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**Data Availability**

ARCTAS-A and -B measurements are available at http://doi.org/10.5067/SUBORBITAL/ARCTAS2008/DATA001, last access 27 April 2020. SEAC$^4$RS measurements are available at http://doi.org/10.5067/Aircraft/SEAC4RS/Aerosol-TraceGas-Cloud, last access 27 April 2020. WINTER measurements are available at https://data.eol.ucar.edu/master_lists/generated/winter/, last access 27 April 2020. ATom-1 and -2 measurements are available at https://doi.org/10.3334/ORNLDAAC/1581, last access 27 April 2020. Ammonia and carbon dioxide measurements from the cabin of the DC-8 are available as an attachment . CSN and CASTNET measurements are available at http://views.cira.colostate.edu/fed/QueryWizard/Default.aspx, last access 27 April 2020. Figures are available at http://cires1.colorado.edu/jimenez/group_pubs.html.

**Competing Interests**

The authors declare no competing interests.

**Author Contribution**

B.A.N., P.C.-J., D.A.D., and J.L.J. designed the experiment and wrote the paper. B.A.N., P.C.-J., D.A.D., H.G., A.V.H., D.P., J.C.S., M.K.S., M.J.C., J.E.D., W.H., and B.B.P. collected and analyzed the data. D.S.J. and A.H. ran GEOS-Chem and provided the output. All authors reviewed the paper.

## Figures

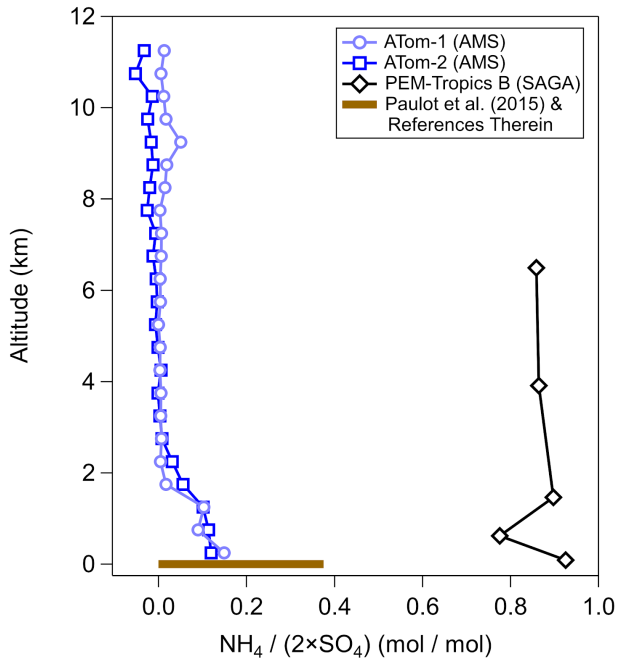


Figure 1. *Vertical profile of sulfate-only ion molar balance (moles($NH_4$)/moles($SO_4$)) measured during PEM-Tropics by collecting the aerosol on filters and analyzing it off-line with ion chromatography (Dibb et al., 2002) and during ATom-1 and -2 by AMS (Hodzic et al., 2020). The ammonium balance profile is for observations collected during ATom-1 and -2 between -20°S and 20°N in the Pacific basin, so that the observations were in a similar location as the PEM-Tropics samples. Also shown is the ammonium balance from observations summarized in Paulot et al. (2015), and reference therein, for the area around the same location as PEM-Tropics.*

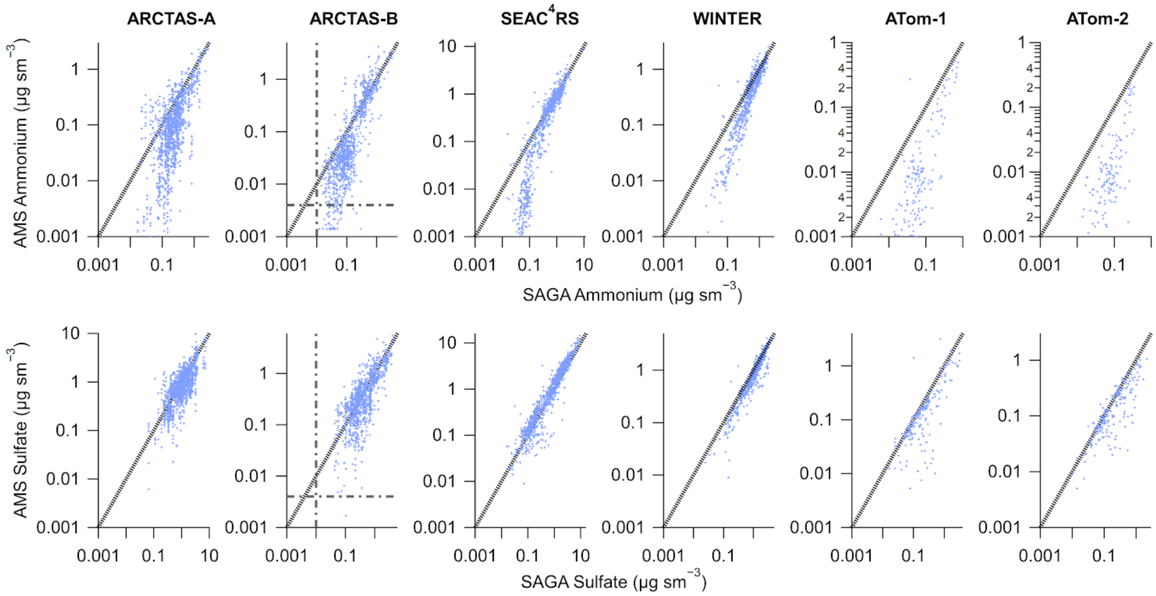


Figure 2. *Scatter plot of AMS (y-axis) versus SAGA filter (x-axis) ammonium (top) and sulfate (bottom) mass concentration from 6 different aircraft campaigns. AMS data have been averaged over the SAGA filter collection times. Black line is the one-to-one line and the grey dash-dot lines are the estimated detection limits for AMS (DeCarlo et al., 2006; Guo et al., 2020) at the SAGA filter collection interval (~5 minutes) and the estimated detection limits for SAGA (Dibb et al., 1999). Data has been averaged to the sampling time of SAGA and has not been filtered for supermicron particles. For ATom-1 and -2, data during ascent and descent has been removed (only level sampling at low altitude and high altitude).*

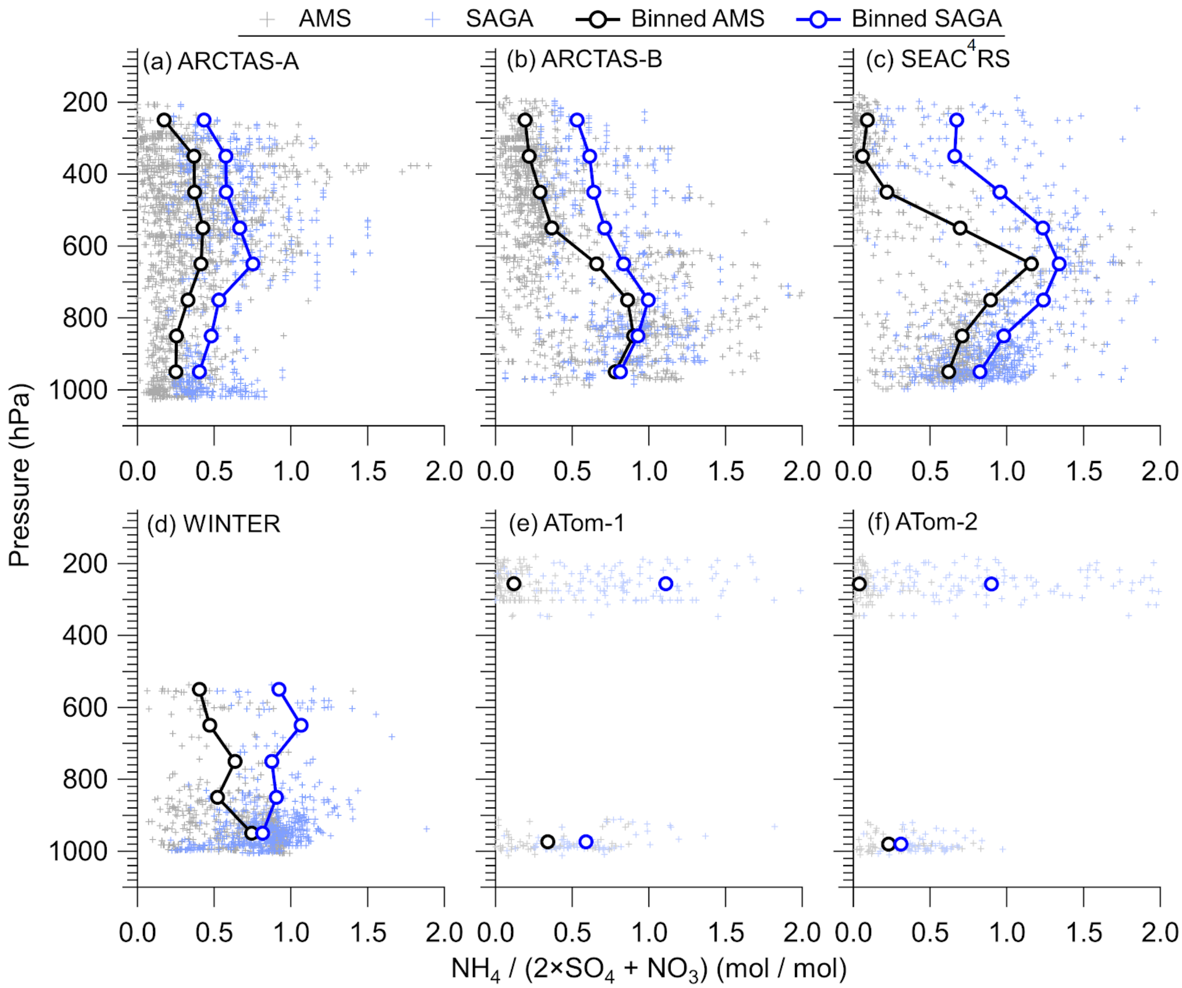


Figure 3. *Vertical profiles of ammonium balance* $((NH_4/18)/(2\times SO_4/96+NO_3/62))$ *for (a) ARCTAS-A, (b) ARCTAS-B, (c) SEAC⁴RS, (d) WINTER, (e) ATom-1, and (f) ATom-2, for AMS and SAGA. The binned data is the mean for each 100 hPa pressure level. The data has been averaged to the sampling time of SAGA filters.*

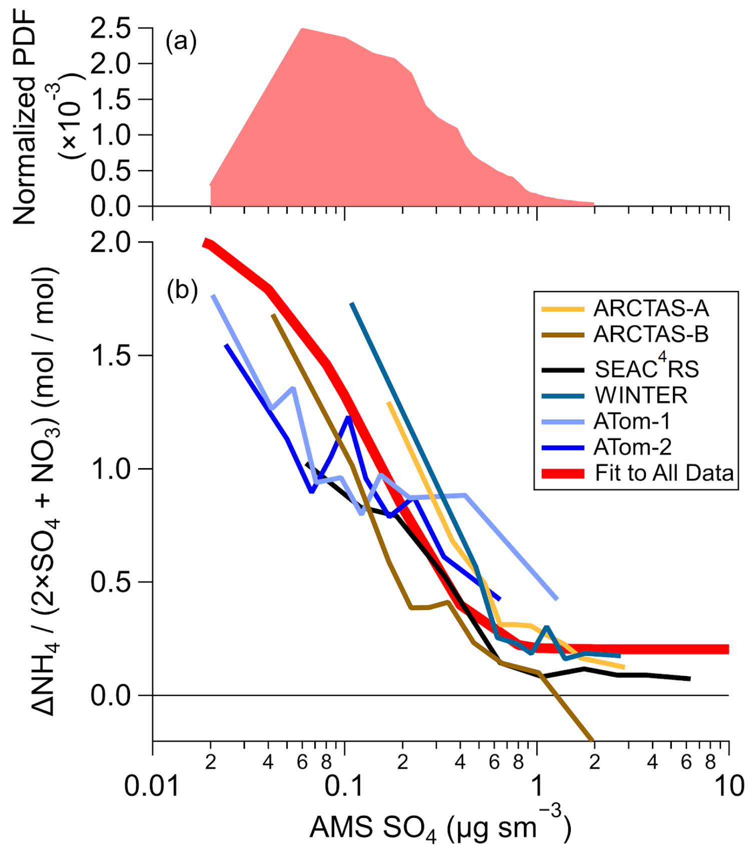


Figure 4. *(a) Predicted normalized probability distribution function (PDF) for tropospheric (pressure > 250 hPa) sulfate from GEOS-Chem for one model year (see SI). (b) Difference between SAGA and AMS ammonium, in mol sm$^{-3}$, divided by AMS sulfate and nitrate, in mol sm$^{-3}$, versus AMS sulfate (μg sm$^{-3}$), for the six different airborne campaigns. The values shown are binned deciles for the five different airborne campaigns. The fit shown in (b) is for all data from all campaigns.*

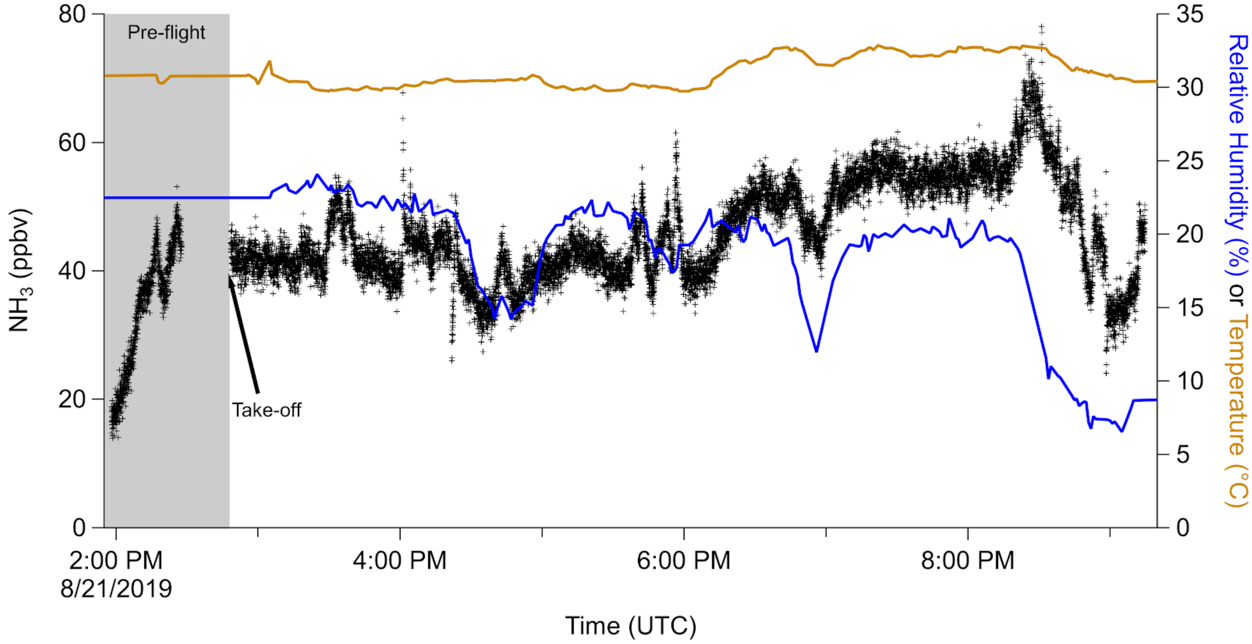


Figure 5. *Time series of ammonia (left) and relative humidity and temperature (right) measured inside the cabin of the NASA DC-8 during a flight during the FIREX-AQ campaign. Time spent prior to take-off is marked with a grey background.*

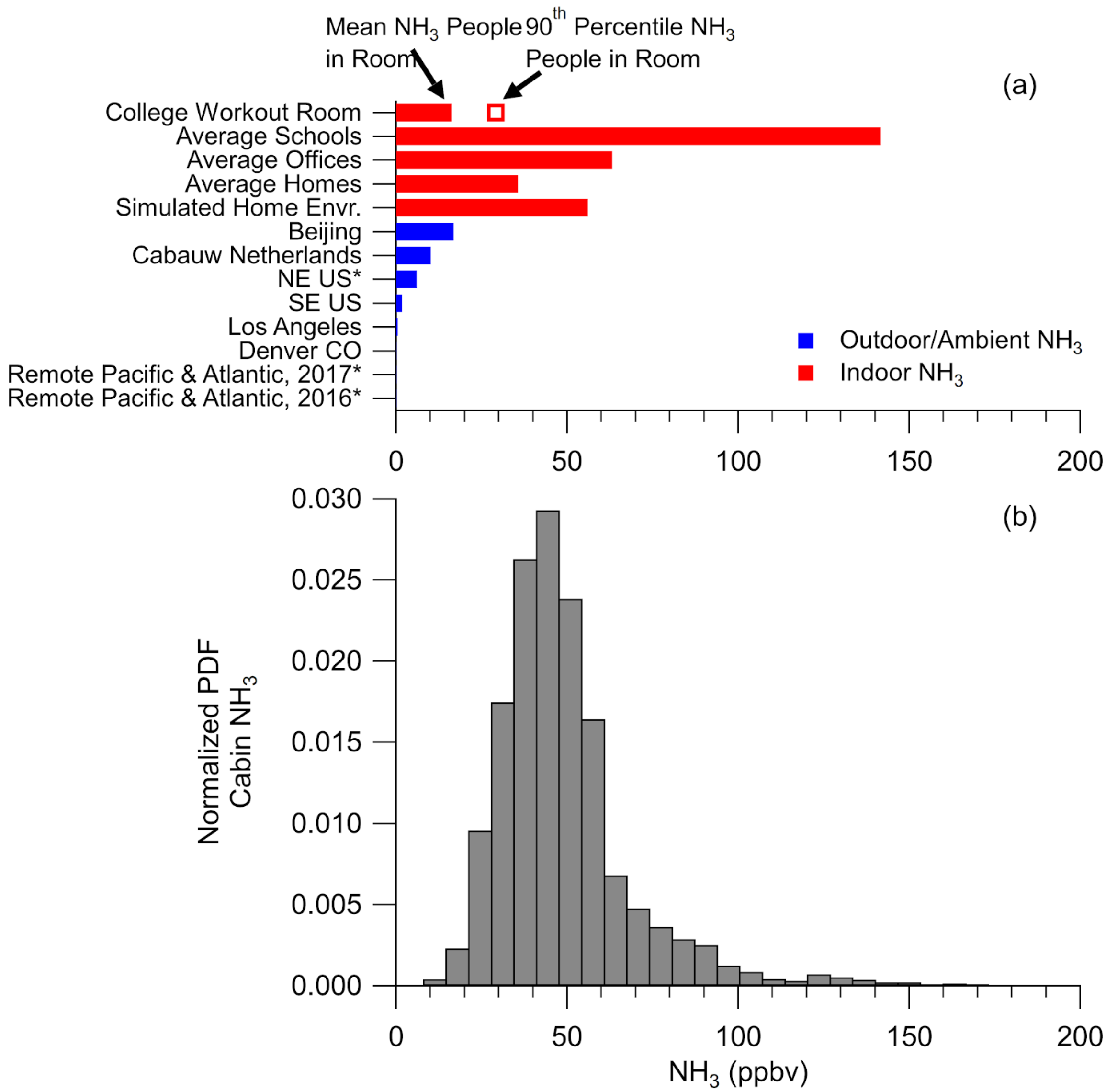


Figure 6. *(a) Ammonia ($NH_3$) (ppbv) reported for studies. See* Table S1 *for references. Asterisk after study name indicates $NH_3$ predicted by thermodynamic model instead of being measured. (b) Normalized probability distribution function (PDF) for $NH_3$, measured in the cabin of the NASA DC-8 during FIREX-AQ.*

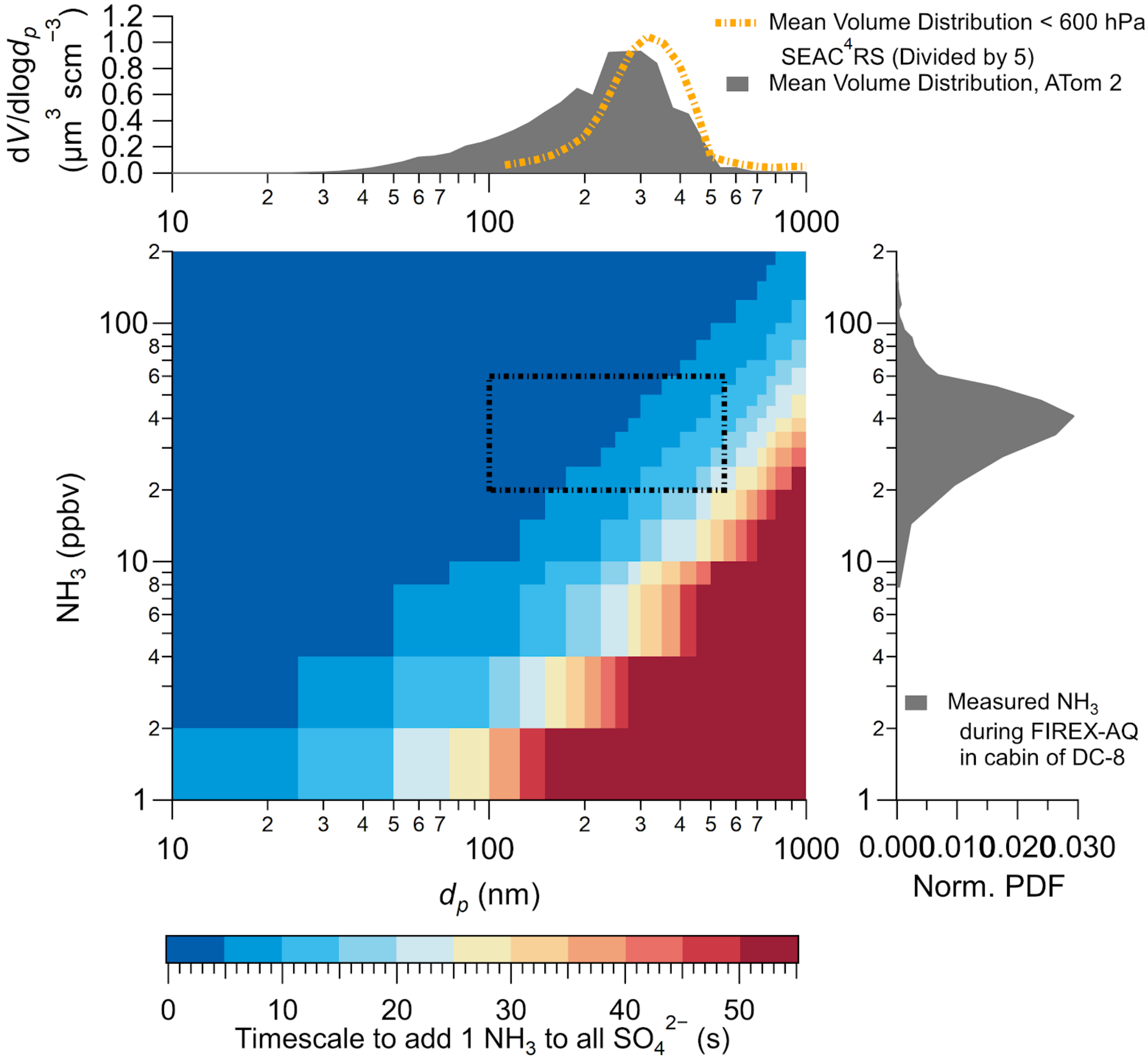


Figure 7. *Theoretical calculation for the amount of time it would take for all the sulfuric acid on the filter to react with one ammonia molecule to become ammonium bisulfate. Volume distribution is the average from SEAC$^4$RS and ATom-2 (adapted from Guo et al. (2020)) and the normalized probability distribution function (Norm. PDF) is from* Fig. 6. *The representative diameter and ammonia mixing ratio are shown as dashed lines in the calculated timescale.*

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