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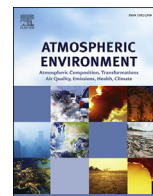2016-07-01

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Short communication

Technical note: An improved approach to determining background aerosol concentrations with PILS sampling on aircraft

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HIGHLIGHTS

- Background distributions of PILS + offline IC measurements do not necessarily follow parametric statistics.
- Histograms of background distributions can be fit to lognormal distributions for accurate background determination.
- This procedure leads to substantially lower calculated limits of detection for ammonium and other inorganic ions.

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ABSTRACT

Particle-into-Liquid Samplers (PILS) have become a standard aerosol collection technique, and are widely used in both ground and aircraft measurements in conjunction with off-line ion chromatography (IC) measurements. Accurate and precise background samples are essential to account for gas-phase components not efficiently removed and any interference in the instrument lines, collection vials or off-line analysis procedures. For aircraft sampling with PILS, backgrounds are typically taken with in-line filters to remove particles prior to sample collection once or twice per flight with more numerous backgrounds taken on the ground. Here, we use data collected during the Front Range Air Pollution and Photochemistry Éxperiment (FRAPPÉ) to demonstrate that not only are multiple background filter samples are essential to attain a representative background, but that the chemical background signals do not follow the Gaussian statistics typically assumed. Instead, the background signals for all chemical components analyzed from 137 background samples (taken from ~78 total sampling hours over 18 flights) follow a log-normal distribution, meaning that the typical approaches of averaging background samples and/or assuming a Gaussian distribution cause an over-estimation of background samples — and thus an underestimation of sample concentrations. Our approach of deriving backgrounds from the peak of the log-normal distribution results in detection limits of 0.25, 0.32, 3.9, 0.17, 0.75 and 0.57 $\mu g\ m^{-3}$ for sub-micron aerosol nitrate (NO_3^-), nitrite (NO_2^-), ammonium (NH_4^+), sulfate (SO_4^{2-}), potassium (K^+) and calcium (Ca^{2+}), respectively. The difference in backgrounds calculated from assuming a Gaussian distribution versus a log-normal distribution were most extreme for NH_4^+, resulting in a background that was 1.58× that determined from fitting a log-normal distribution.

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1. Introduction

The composition of atmospheric aerosols is the result of primary emissions, secondary aerosol formation, and atmospheric aging chemistry. Atmospheric chemists have approached measurements of the aerosol composition using both single particle and bulk property analysis (Farmer and Jimenez, 2010). One method for characterizing bulk aerosol composition is collection of aerosols into the aqueous-phase using a Particle-into-Liquid Sampler (PILS). These systems have been used extensively over the last decade in both aircraft and ground based studies, and have been used to

study secondary aerosol formation (Weber et al., 2007) and carbonaceous water-soluble organic aerosols (Sullivan et al., 2006). Samples collected by PILS can be analyzed on-line, or collected for separate off-line analysis with ion chromatography (IC) or other aqueous analyses. Aircraft represent a particularly challenging platform for aerosol measurements due to the rapidly changing pressure, differences in pressure and temperature between inlet and instrument, and the need for high time resolution to capture rapidly changing ambient concentrations (Farmer and Jimenez, 2010). Due to timing and weight issues, off-line analyses are often used for aerosol chemical quantification by PILS in aircraft sampling (Sorooshian et al., 2006).

The application of PILS coupled with IC work for aircraft measurements was first described by Orsini et al. (2003). This work described the LOD (limit of detection) of the instrument as depending on the noise in the baseline of the chromatograms — specifically, the running conditions of the IC including column and eluant conditions, temperature and quality of deionized water. Orsini et al. (2003) determined the 'blank' from samples collected after air had passed through a filter that was placed upstream of gas removal denuders. LODs were determined as the average filter signal plus 3σ, where σ is the standard deviation of the chromatogram baseline. These LODs ranged from 0.002 and 0.004 $\mu g \ m^{-3}$ for SO_4^{2-} and NO_3^-, respectively, to 0.059 and 0.288 $\mu g \ m^{-3}$ for NH_4^+ and K^+, respectively. Sorooshian et al. (2006) similarly collected samples with PILS for subsequent off-line IC analysis, using a filter to collected background samples; the authors noted that the background was affected not only by the IC system, but also by contamination in the washflow water, plumbing in the PILS, both PILS collection and IC vials, and sample transfer between vials. In laboratory characterization, the background was described as the mean $+3\sigma$, where σ is the standard deviation of the background concentrations determined from $n > 300$ filter samples. These background levels ranged from 0.10 and 0.11 $\mu g \ m^{-3}$ for SO_4^{2-} and NO_3^- to 0.27 and 0.17 $\mu g \ m^{-3}$ for NH_4^+ and K^+, respectively. Sorooshian et al. (2006) further note that in this system, the IC system typically accounts for <0.05 $\mu g \ m^{-3}$ for most ions. Thus, characterizing the true background of the entire PILS sample collection and analysis is essential for accurate aerosol composition measurements.

The extent to which laboratory characterization of the background represents atmospheric samples has not been fully explored for the PILS + offline IC sampling system. PILS collects samples on the timescale of minutes, and collecting multiple background samples during an aircraft flight can substantially reduce the number of ambient samples collected, thus affecting regional coverage of aerosol composition. Even instruments with fast time resolution (1 Hz) aboard an aircraft travelling at 50 m s^{-1} provide a 50 m spatial average, a scale larger than most pollution plumes (Farmer and Jimenez, 2010). This trade-off in time collecting background versus ambient samples has resulted in PILS background measurements typically being limited to one to two samples during a flight. Additional samples may be taken on the ground before takeoff or after landing. However, as changes in altitude, temperature, pressure, etc. can potentially alter instrument backgrounds during flight, it is unclear if the current procedures for aircraft PILS sampling adequately capture a representative background.

The use of air filtered samples to determine the background is a matrix-specific method of determining backgrounds and LODs. Background filter samples should account for gas-phase interferences that were not removed by the denuders, and re-equilibration between the gas and liquid phases in the tubing between the denuders and the condensation chamber (Sullivan et al., 2006; Weber et al., 2007). Other potential interferences in the

sampling system include evaporation of semivolatile species in the inertial impactor, condensation chamber, or sampling lines; formation of large droplets at high steam tip temperatures and subsequent losses; inadequate cleaning of the surface of the droplet impactor; and changes in inlet transmission efficiency due to changes in pressure.

Here we investigate the variability in background concentrations collected during an aircraft campaign with the aim of improving the analytical methodology for aircraft sampling of aerosol composition using a PILS coupled to an off-line sampler. We suggest an approach of fitting the distribution of background sample concentrations to a lognormal distribution in order to find the mode, which more accurately represents the background concentration.

2. Materials and methods

2.1. Research flights

The Front Range Air Pollution and Photochemistry Éxperiment (FRAPPÉ) included 18 research flights between July 26 and August 18, 2014, totaling ~78 sampling hours, on the National Center for Atmospheric Research (NCAR) C-130 aircraft from Rocky Mountain Metropolitan Airport in Broomfield, CO. The majority of flights took place during the day in sunny conditions, predominantly at altitudes between ~1200 and 7500 m. Flight paths were designed to characterize the chemical background in the Colorado Front Range and to quantify pollution sources in the region.

2.2. PILS collection

Aerosol samples were collected using a Brechtel Manufacturing, Inc. model 4001 PILS system with an 80 position carousel auto-collector (Sorooshian et al., 2006). The aircraft inlet used for this study was a submicron aerosol inlet (SMAI) (Craig et al., 2014; Craig et al., 2013; Moharreri et al., 2014). A detailed explanation of PILS collection is described in detail elsewhere (Orsini et al., 2003; Sorooshian et al., 2006). Briefly, ambient particles were pulled through the sample inlet (~15 LPM, inlet ~4.3 m long with ~4.8 mm average inner diameter), a single stage impactor (size cut 1.0 μm), and sodium carbonate and phosphorous acid coated denuders (residence time between denuder and condensation chamber ~0.08 s; between inlet and condensation chamber ~1.2 s) before entering the PILS instrument where the ambient air is rapidly mixed with steam from a steam generator. The steam tip in the PILS instrument was maintained at 100 ± 5 °C. As the ambient air and steam mix, the steam cools, leading to a supersaturated environment within the condensation chamber. Aerosol particles grow into droplets between 1 and 5 μm in diameter. These droplets are collected on an impaction plate. A wash flow solution (flow rate ~1.7 mL min^{-1}) containing high purity water (18 MΩ) and 10 μM LiBr (lithium bromide) is then directed across the impaction plate, transporting the droplets to polypropylene collection vials (MicroSolv, 9502S-PP-CLEAR) over a period of 5 min per sampling event. Liquid flows were controlled by a peristaltic pump. Due to space and logistical limitations, only one sampling tray was allowed per flight, limiting the number of samples that could be collected in flight. This limited the sampling time and thus the number of in-flight background samples, resulting in the majority of background samples being taken either immediately before or after the actual flight.

To determine the background of the entire system, a manual valve was placed downstream of the denuders to direct sample flow through a HEPA filter before entering the PILS condensation chamber, similar to the setup described in Sorooshian et al. (2006).

Hereon, samples collected after passing through the filter are referred to as 'background samples', while those that did not pass through the filter are referred to simply as 'ambient samples'. This valve switch resulted in a slight pressure difference at the entrance to the PILS system, enabling exclusion of any filtered samples that included ambient air from the background analysis. Three to nine background vials were collected at both the beginning and end of each flight (6 + background samples per flight). We note that in order to minimize the number of mixed ambient and background vials, the valve switch was timed to occur at the end of an ambient sample vials as the auto-collector switched to a new vial, as indicated by the BMI-Auto Collector v1.0.vi software accompanying the PILS.

Following landing, the vials were removed from the carousel and the slit caps were replaced with solid caps for storage. Vials were stored and transported on ice in sealed, insulated containers; vials were stored in the lab at 4 °C until analyzed. IC analysis was performed on each sample within 48 h of the corresponding flight, typically <24 h.

2.3. IC analysis

We used a Dionex ICS-3000 ion chromatograph with conductivity detection and an AS50 autosampler to analyze the background and ambient samples for inorganic ions. A 200 μL injection loop was employed; temperature was maintained at 30 °C. Elution was carried out isocratically with 17 min run times. The IC was calibrated for cations (Li^+, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) and anions (F^-, Cl^-, NO_2^-, Br^-, NO_3^-, PO_4^{3-}, and SO_4^{2-}). No co-elution of these reported species was observed during the analysis. IC background concentrations for each ion were accounted for by running vials containing only high purity water (18 MΩ) — two background water samples before the sample vials, and one afterwards. The IC background for each ion was determined as the average of these three samples and was subsequently subtracted from the ion signals for every sample and background sample vial. Analyte peaks were integrated using the Chromeleon software package.

A Dionex IonPac CS12A (150 mm × 3 mm i.d.) column was used for cation analysis, with a Dionex IonPac CG12A (30 mm × 3 mm i.d.) guard column. We used a Thermo Scientific Dionex 2 mm CERS 500 electrolytically regenerated suppressor, operated at 40 mA. The eluent was 20 mM methanesulfonic acid (MSA) for cation analysis and the flow rate was 0.50 mL min^{-1}.

Anion analysis used a Dionex IonPac AS14A (250 mm × 4 mm i.d.) column, a Dionex IonPac AG14A (50 mm × 4 mm i.d.) guard column, and a Thermo Scientific Dionex 4 mm AERS 500 electrolytically regenerated suppressor, operated at 50 mA. The eluent was 8 mM sodium carbonate and 1 mM sodium bicarbonate and the flow rate was 1.0 mL min^{-1}.

It is important to note that there are two backgrounds that are referred to herein: the IC backgrounds (18 MΩ high purity water) and the PILS/sampling backgrounds (HEPA filter, background vials transported and treated the same way as sample vials). All samples, including background filter samples, have been IC background subtracted, and are calibrated for IC response. Additionally, all reported values are flow corrected using the dilution factors obtained from the LiBr wash flow solution flow rate and inlet flow rate of the PILS, resulting in concentrations reported in units of μg m^{-3}.

3. Results

We focus on eight inorganic ions: Cl^-, NO_2^-, NO_3^-, SO_4^{2-}, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}. Of 980 vials collected, 836 contained ambient sample and 137 contained filter background only. Inlet pressure data was used to identify vials that included mixed ambient/

background samples, resulting in the exclusion of 7 vials from the following analysis. The distribution of background measurements for each ion, in units of air-equivalent concentration, can be described by a histogram. We fit these histograms using both lognormal and Gaussian distributions. The lognormal fit followed the general equation

$$y = y_o + Ae^{-\left[\frac{\ln\left(\frac{x}{x_o}\right)}{width}\right]^2} \tag{1}$$

where x_o and $width$ represent the mode and standard deviation, respectively, and y_o and A are additional fit parameters. The Gaussian fit followed the general equation

$$y = y_o + Ae^{-\left[\frac{x - x_o}{width}\right]^2} \tag{2}$$

where parameters are similar to E1, although x_o is taken to represent the mean of the population. Table 1 compares these fit parameters to the mean and standard deviation determined directly from the dataset. A single background concentration for each ion was determined from the mode or mean of the corresponding histogram; this background concentration is then subtracted from every ambient data point. The limits of detection (LOD, S/N = 3) for each ion were determined as 3σ for each distribution (Table 1). This approach to backgrounds assumes that the background was constant for each ion throughout the 3 week campaign, consistent with the observed distribution. This background concentration was subtracted from each ambient sample vial. Statistics for the resulting ambient sample dataset are summarized in Table 2.

4. Discussion

Background filter data are typically averaged to derive a background, which is then subtracted from all ambient sampling data to derive aerosol concentrations. However, we note that this approach assumes that the data population follows a Gaussian distribution, and that data points are evenly distributed around the upper and lower sides of the mean. These assumptions are not met in this dataset: the mean calculated from the data shows high background averages calculated as the mean of all background data relative to the mean calculated from a Gaussian distribution. Thus fitting the data to a non-parametric distribution, such as the lognormal distribution used in this manuscript, avoids the pitfalls of assuming that the dataset has an equal number of points above and below the mean. The data distributions shown in Fig. 1 suggest that the background samples represent a single population that is reasonably well-described by either a Gaussian or lognormal distribution. However, the lognormal distribution accounts for the observed positive tail, which is common when performing analyses via chromatographic techniques, as these techniques result only in a positive measurement — i.e., peak areas can only be ≥zero. The background of the samples are best described as the mode of the distribution, calculated from the peak of the lognormal fit. Consistent with the idea that the filter measurements are subsamples from a single population, backgrounds for each ion are poorly correlated with altitude, steam tip temperature, ambient temperature or ambient pressure ($r^2 < 0.3$ for all ions). This suggests that backgrounds are not consistently dependent on instrument parameters or external atmospheric variables. No statistically significant difference was found between background measurements taken at the beginning versus end of each flight, nor between background measurements taken between different flights,

Table 1
Summary of background sample chemical analysis, including means, modes and standard deviations (σ) derived directly from the data and from Gaussian and lognormal fits (n = 137). The LOD is presented as 3σ for each approach to determine the width of the distribution. All values listed were corrected for ion chromatography calibrations and backgrounds, and flow corrected using the dilution factors calculated for each background sample based on the flow rates of the PILS inlet and wash flow solution. All data are presented in air equivalent concentrations of μg m^{-3}. Uncertainties for all fit parameters are reported in brackets.

	Directly calculated			Gaussian fit			Lognormal fit		
	Average (μg m^{-3})	Standard deviation, σ	3σ (LOD)	Mean (μg m^{-3})	σ (fit width)	3σ (LOD)	Mode (μg m^{-3})	σ (fit width)	3σ (LOD)
Cl$^-$	1.46	2.07	6.21	0.85 (0.02)	0.35 (0.03)	1.05	0.79 (0.02)	0.43 (0.04)	1.29
NO$_2^-$	1.7	0.31	0.93	1.624 (0.004)	0.170 (0.006)	0.51	1.617 (0.004)	0.106 (0.004)	0.32
NO$_3^-$	3.5	0.67	2.01	3.14 (0.01)	0.083 (0.006)	0.25	3.14 (0.01)	0.083 (0.006)	0.25
SO$_4^{2-}$	5.6	0.27	0.81	5.574 (0.008)	0.31 (0.01)	0.93	5.568 (0.008)	0.056 (0.002)	0.17
NH$_4^+$	0.97	0.73	2.19	0.63 (0.06)	0.81 (0.1)	2.43	0.40 (0.04)	1.30 (0.17)	3.90
K$^+$	1.22	1.25	3.75	0.898 (0.008)	0.21 (0.01)	0.63	0.878 (0.007)	0.25 (0.01)	0.75
Mg^{2+}	0.86	0.30	0.90	0.734 (0.005)	0.149 (0.007)	0.45	0.724 (0.004)	0.217 (0.009)	0.65
Ca^{2+}	1.70	0.63	1.89	1.32 (0.01)	0.24 (0.02)	0.72	1.31 (0.01)	0.19 (0.01)	0.57

Table 2
Summary of aerosol sample composition for (n = 54) sample measurements from a single research flight (RF 06, 08/02/2014). All values are reported in μg m^{-3}. Each value has been IC background subtracted (18 MΩ high purity water) and calibrated, in addition to background subtracted (mode of background sample data, calculated from lognormal distribution).

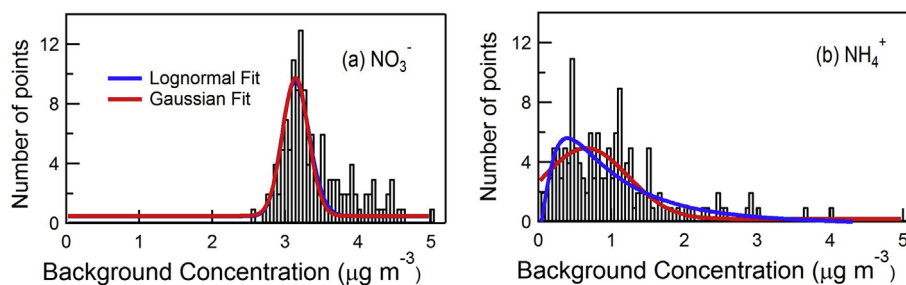| Ion	#Data points > LOD$_{lognormal}$	Average sample concentration (μg m^{-3})	Standard deviation, σ (μg m^{-3})	Maximum sample concentration (μg m^{-3})	Minimum sample concentration (μg m^{-3})
Cl$^-$	0	0.28	0.1	0.62	0.15
NO$_2^-$	54	0.43	0.03	0.52	0.38
NO$_3^-$	54	0.85	0.08	1.03	0.72
SO$_4^{2-}$	54	1.45	0.08	1.77	1.38
NH$_4^+$	0	0.65	0.22	0.99	0.22
K$^+$	1	0.27	0.11	0.19	0.83
Mg^{2+}	0	0.21	0.06	0.58	0.15
Ca^{2+}	2	0.43	0.29	2.48	0.31

Fig. 1. Histograms of the background concentration (μg m^{-3}) for (a) NO$_3^-$ and (b) NH$_4^+$ PILS-IC aircraft data from the 2014 FRAPPÉ campaign. The distributions are fit with both lognormal (blue) and Gaussian (red) fits. The typical approach of averaging a few background samples could lead to overestimation of the background concentrations and LODs, while the use of a histogram and its respective best fit better captures variability in the backgrounds. The number of bins used for both histograms was 100, and the bin size was 0.05 μg m^{-3}. No data points are excluded from the figures. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this article.)

suggesting that the sampling system (PILS + IC) is robust, albeit with a substantial variance in the background concentrations for inorganic ions.

It is apparent from Table 1 that merely averaging the background dataset results in an overestimate of the background. Averaging 1–2 filter samples to determine a background for each flight may not accurately represent the background for PILS-IC sampling; instead, multiple filter samples are required to accurately describe the population of background samples. More importantly, merely calculating the average and standard deviation of the background samples does not necessarily capture the background distribution due to the positive tail observed in the background distributions. Fitting the data to a parametric or non-parametric distribution is essential for accurate background measurement. For example, the background determined by a simple average of the dataset is 2.4 times the background determined by the lognormal distribution for ammonium, though the two approaches are within uncertainty for sulfate. The overestimation of

the ammonium background could affect calculations of aerosol pH, mass and ion balance.

Accurate calculation of the standard deviation of the background samples, and thus LODs, is essential for maintaining data quality. The use of a lognormal fit to derive the width of the background distribution and thus the LOD (=3σ$_{lognormal}$) provided substantially lower detection limits than those determined from calculating the standard deviation directly from the data, or even from a Gaussian fit of the data. This is due to the uneven distribution, and confirms that the data does not follow traditional parametric statistics. For example, the LOD for nitrite determined from the calculated versus Gaussian fit standard deviation of the blanks was 0.93 versus 0.51 μg m^{-3}, as opposed to the LOD of 0.32 μg m^{-3} determined from the lognormal fit of the histogram of filter samples. As every sample was determined to have a nitrite concentration between 0.38 and 0.52 μg m^{-3} (Table 2), incorrect determination of the detection limit reflects the difference between every data point being considered above or below the detection

limit.

The dataset presented herein suggests that background data collected by PILS-IC should be fit to both parametric and non-parametric distributions to determine whether the assumptions of a Gaussian distribution are met and to accurately identify the true mean (or mode) and standard deviation of the dataset. While enough background samples must be taken to determine the background distribution (ideally ≥30), and thus background concentrations and distribution widths, this work suggests that in-flight background samples, while ideal, may not be necessary for this type of sampling. However, we note that the aircraft sampling described in this campaign covered a very limited altitude range, and that correlations between backgrounds and ambient conditions must be verified for other instruments and flight conditions For longer campaigns, backgrounds may increase over time as, for example, denuder efficiency decreases. Ground-based campaigns are subject to less vibration, smaller changes in pressure and temperature, and less cycling of power on the instrument, and are therefore expected to show greater precision and lower LODs. Background sampling in such campaigns is also expected to be more frequent. However, we encourage researchers to consider using histograms of their background concentrations to determine whether the average of a few points, or interpolation between individual background points, is the ideal approach. We note that while the lognormal distribution was determined to be the most appropriate fit for this dataset, backgrounds from other datasets may be more accurately represented by different distribution functions.

5. Conclusions

This study suggests that the background distribution of many ions detected by the PILS + offline IC method, including ammonium, do not follow a parametric distribution. Thus, the current methodology for determining background concentrations for aircraft-based PILS measurements coupled to off-line analysis from calculated averages and standard deviations of background samples may be insufficient to accurately capture the background population, and may lead to an underestimate of sample concentration and overestimate of instrument detection limits. Instead,

we find that fitting the data to a lognormal distribution and using the mode and standard deviation determined from that fit to calculate the background and detection limit better represents the instrument background.

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