Assessment of Ethanol Trends on the ISS

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The International Space Station (ISS) Environmental Control and Life Support System (ECLSS) provides a working environment for six crewmembers through atmosphere revitalization and water recovery systems. In the last year, elevated ethanol levels have presented a unique challenge for the ISS ECLSS. Ethanol is monitored on the ISS by the Air Quality Monitor (AQM). The source of this increase is currently unknown. This paper documents the credible sources for the increased ethanol concentration, the monitoring provided by the AQM, and the impact on the atmosphere revitalization and water recovery systems.

Nomenclature

AQM	=	Air Quality Monitor
BMP	=	Russian Micropurification Unit
CCAA	=	Common Cabin Air Assembly
CDRA	=	Carbon Dioxide Removal Assembly
COA	=	Catalytic Oxidizer Assembly
DMSD	=	dimethylsilanediol
GC	=	Gas Chromatograph
GSC	=	Grab Sample Container
GUI	=	Graphical User Interface
ISS	=	International Space Station
JAXA	=	Japan Aerospace Exploration Agency
MART	=	Multilateral Anomaly Resolution Team
M&P	=	Materials and Processes
RHS	=	Reactor Health Sensor
SKV	=	Russian condensing heat exchanger assembly
TCC	=	trace contaminant control
TCCS	=	Trace Contaminant Control Subassembly
TEC	=	Toxicology and Environmental Chemistry
TOC	=	total organic carbon
UPA	=	Urine Processor Assembly

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VOC	=	volatile organic compound
VUA	=	Volatile Usage Agreement
WPA	=	Water Processor Assembly
A	=	bulk process gas molar flow rate
atm	=	atmospheres
cm	=	centimeter
g	=	gram
h	=	hour
k_H	=	Henry's Law constant
kPa	=	kilopascal
L	=	condensate molar flow rate
mg	=	milligram
m	=	meter
p	=	partial pressure
P	=	atmospheric pressure
Pa	=	pascal
x	=	liquid phase mole fraction
y	=	gas phase mole fraction

I. Introduction

THE Atmosphere Revitalization System (ARS) and Water Recovery System (WRS) maintain a working environment for six crew on the International Space Station (ISS). These systems were designed based on specific interfaces, including the influent contaminant load to be processed and the air and water quality required to ensure crew health. Strict controls are implemented on the ground to insure materials transported to ISS do not impact the performance of the ARS and WRS. Furthermore, air and water quality are monitored both aboard ISS and via analysis of samples returned to the ground. Excursions in the specified contaminant load can have different impacts to the ARS and WRS. In some cases, the impact is negligible if the excursion is well within the system capacity. In other cases, a significant increase in contaminant load can have a significant impact to system performance, resulting in increased consumable rates or the inability of the system to meet the specified air or water quality requirements. In 2015, a significant sustained increase in atmospheric ethanol levels had a significant impact on WRS performance by exceeding system capacity. This paper discusses the ethanol trend on ISS in 2015, how ethanol is treated by the ARS and its impact on WRS performance, and credible sources of ethanol on ISS.

II. Ethanol Sources and Removal aboard the ISS

Ethanol is a pervasive, commonly observed chemical contaminant in spacecraft cabin atmospheres. Alcohols have historically accounted for nearly 80% of the non-methane volatile organic compound (VOC) concentration in the ISS cabin with a total concentration averaging ~5.5 mg/m^3.[1] Ethanol typically accounts for >70% of the total alcohol concentration. Chemical and physical property data summarized by Table 1 serve as the basis for trace contaminant control and cabin mass balance calculations as well as for evaluating the mass transport of ethanol into humidity condensate collected by the ISS condensing heat exchanger units.

Table 1. Ethanol chemical and physical properties.

PROPERTY	VALUE
CAS Number	65-17-5
Molecular weight (g/mole)[2]	46.07
Phase	Liquid
Color[1]	Colorless
Odor[1]	Ethereal, vinous
Boiling point (ºC)[1]	78.37
Liquid density at 25 ºC (g/cm^3)[2]	0.789
Critical temperature (°C)[2]	243
Critical pressure (kPa)[2]	6383
Critical volume (cm^3/mole)[2]	167
Critical compressibility[2]	0.248
Henry's Law constant, k_H (mole/m^3-Pa)[3]	1.712
Henry's constant temperature dependence factor, $d\ln(k_H)/d(1/T)$[4]	7116.76
Solubility in water[1]	Miscible
Vapor pressure at 20 ºC (kPa)[1]	5733

1. G.G. Hawley. The Condensed Chemical Dictionary. Tenth Edition. 1981.
2. R.C. Reid, J.M. Prausnitz, and T.K. Sherwood. The Properties of Gases and Liquids. Third Edition. 1977.
3. Average from Sander, R., Compilation of Henry's Law Constants V.3.99, 28 November 2014.
4. Average temperature dependence factor from Sander, $k_H(T)=k_H \times e^{\{d\ln(kH)/d(1/T)[1/T-1/298.15]\}}$

A. Primary Ethanol Generation Sources under Normal Conditions

Ethanol generation sources include equipment offgassing, crew metabolism, payload operations, and a myriad of personal care products. The ethanol generation load for trace contaminant control design consistent with the upper bound of the 95% confidence interval consists of equipment offgassing at 0.00785 mg/day-kg and a crew metabolic load of 4.26 mg/day-person.[2] For a vehicle the size of the ISS, the estimated equipment mass contributing to equipment offgassing, assuming 150 kg/m^3 of cabin volume, is ~954 mg/day. For a crew of six, the daily production is ~26 mg/day. In total the basic production rate excluding payload operations and crew activities is ~980 mg/day (~0.98 g/day). Using the 1-year period between 5 March 2013 and 3 March 2014 as a basis, the 95% confidence interval for the ISS cabin ethanol concentration ranged between 2.90 mg/m^3 and 3.54 mg/m^3. During this period the average concentration reported in thirty-six cabin grab sample analyses was 3.22 mg/m^3 with a standard deviation of 0.99 mg/m^3. Variation may result due to crew activities and vehicle docking activities. Typically concentration transients from such activities are short-lived. Using the 95% confidence interval range boundaries of 2.90 mg/m^3 and 3.22 mg/m^3 to address typical concentration variation, a cabin mass balance accounting for the primary removal mechanisms indicates total ethanol generation between ~1.9 g/day and ~2.4 g/day. At this rate, the contribution from payload operations and crew activities is estimated to typically contribute between ~0.96 g/day and ~1.39 g/day.

B. Ethanol Removal Mechanisms and Phenomena

Ethanol is removed primarily by trace contaminant control (TCC) and humidity control equipment. Aboard the ISS, this equipment consists of the U.S. Trace Contaminant Control System (TCCS), the Russian Micropurification Unit (BMP), the U.S. Common Cabin Air Assembly (CCAA) and the Russian humidity control assembly (SKV). While temporary removal by the U.S. Carbon Dioxide Removal Assembly (CDRA) can occur, a large portion of the ethanol adsorbed by CDRA is typically returned to the cabin during desiccant bed regeneration yielding a negligible net removal capability compared to that provided by the combined action of the TCC and humidity control equipment.[3-5] The specific characteristics of ethanol removal by the TCC and humidity control equipment aboard the ISS are presented by the following discussion.

1. Removal by Trace Contaminant Control Processes

The TCCS purifies the cabin atmosphere by flowing process air through activated carbon and a thermal catalytic oxidation reactor. The total flow of 15.3 m^3/h is processed by the activated carbon bed after which 4.6 m^3/h is processed through the catalytic oxidation reactor. Details on the TCCS are found in Ref. 6. The activated carbon bed in the TCCS is not regenerated. Testing and engineering analysis have shown that ethanol begins to break through the carbon bed within several months of installation. This leaves the catalytic oxidizer assembly (COA) as the primary means to remove ethanol. At the COA's operating conditions the removal efficiency is 100% per single pass. This yields a typical effective removal flow of 4.6 m^3/h.

The BMP flow rate is approximately 25 m^3/h through an expendable fixed activated carbon bed, two regenerable activated carbon beds, and an ambient carbon monoxide oxidation reactor. Details on the BMP are found in Ref. 7. Testing conducted in 1997 showed the net ethanol removal efficiency declining from 100% to 40% over the 20-day period between bed regeneration.[8,9] Therefore, the average efficiency provided by the BMP is assumed to be 70% yielding an effective removal flow of 17.5 m^3/h.

2. Removal by Humidity Control Processes

Trace contaminant removal from air via absorption by humidity condensate is a process that follows Henry's Law as defined by Eq. 1.

$$p = k_H x \tag{1}$$

In this equation, p is the partial pressure of the chemical contaminant in the bulk gas phase, k_H is the Henry's Law constant, and x is the mole fraction of the contaminant in the bulk liquid phase. The units for p and k_H are atm and atm/mole fraction, respectively.

Mass balance equations for co-current absorption have been developed for a typical spacecraft condensing heat exchanger.[10] Using Henry's Law as the equilibrium condition, the solved mass balance equation relates liquid phase mole fraction and gas phase mole fraction according to Eq. 2.

$$x = y/(L/A + k_H/P) \tag{2}$$

In Eq. 2, x is the liquid phase mole fraction, y is the gas phase mole fraction, L is the condensate collection molar flow rate (moles/h), A is the bulk process air molar flow rate (moles/h), H is the Henry's Law constant in atm/mole fraction adjusted for the condensing heat exchanger temperature, and P is the total cabin pressure in atm. Equation 2 can be used as the basis for calculating bulk gas phase and liquid phase portioning. Figure 1 shows that pairs of measured

cabin atmosphere concentrations and humidity condensate loading is within the range expected by Henry's Law as predicted by Eq. 2.

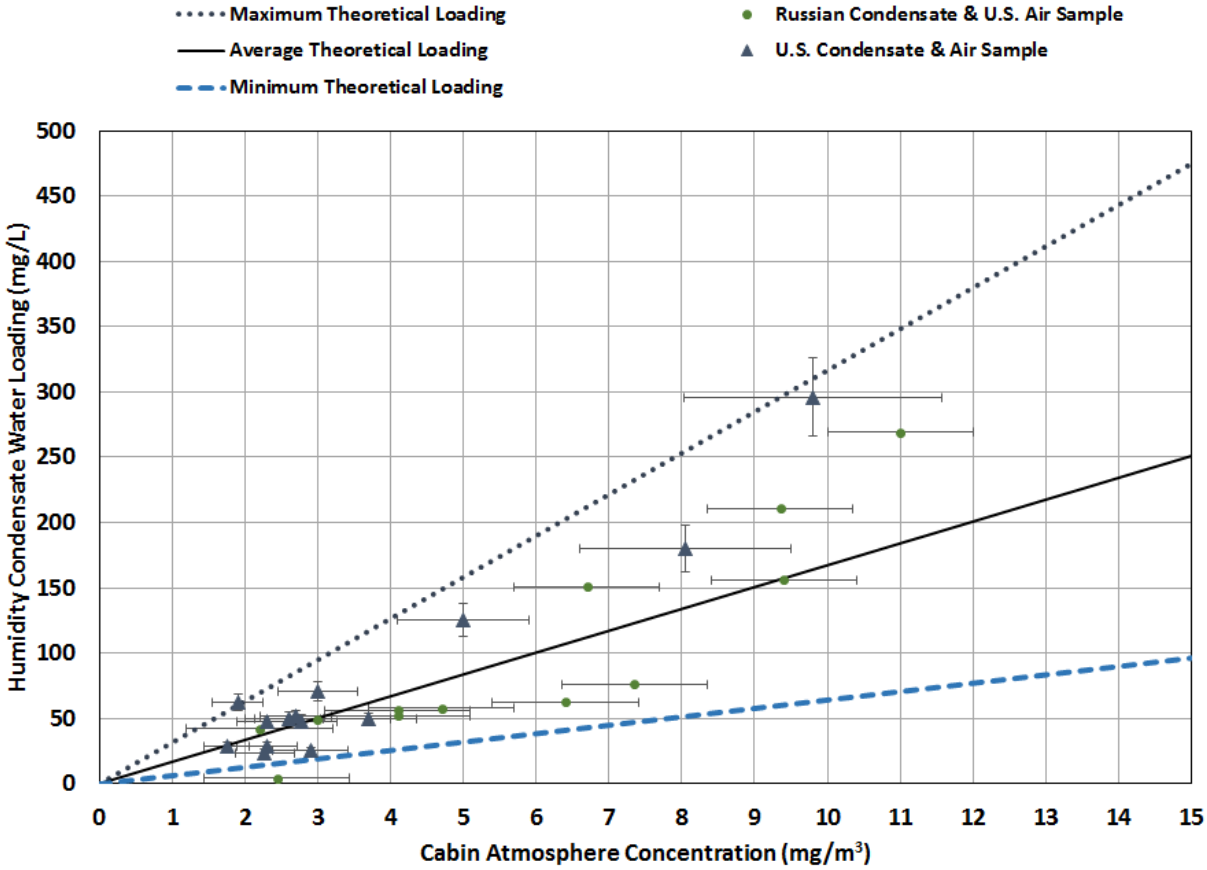

Figure 1. The relationship between cabin atmospheric concentration and humidity condensate loading. *The measured sample pairs fall within the expectation for the range of Henry's Law constants reported in the literature.*

Further evaluation using Eq. 2 as the basis calculates the single pass removal efficiencies summarized by Table 2 for a CCAA operating temperature of 4 °C and an SKV operating temperature of 14 °C. The process air flow through the SKV condensing heat exchanger core is 144 m^3/h. At a typical 50% bypass, the process air flow through the CCAA condensing heat exchanger core is 339.8 m^3/h. The humidity condensate collection is typically 67% by the SKV and 33% by the CCAA. Under this operating condition the SKV removes up to a 4-crewmember equivalent latent load while the CCAA removes a 2-crewmember equivalent latent load. Under these flow and temperature conditions the effective removal flow for the CCAA and SKV is ~2.72 m^3/h and ~3.02 m^3/h.

Table 2. Calculated ethanol removal efficiency and effective flows for the CCAA and SKV.

Crew Equivalent*	Condensate Collection Rate (kg/h)	CCAA Efficiency (%)	CCAA Effective Flow (m^3/h)	SKV Efficiency (%)	SKV Effective Flow (m^3/h)
1	0.0938	0.40	1.35	0.53	0.76
2	0.1875	0.80	2.72	1.06	1.53
3	0.2813	1.19	4.04	1.58	2.28
4	0.3750	1.58	5.37	2.10	3.02
5	0.4688	1.97	6.69	2.61	3.76
6	0.5625	2.35	7.98	3.11	4.48

**1.4 kg/person-day for 8 hours sleep and 16 hours normal activity. Derived from Human Integration Design Handbook, NASA SP-2010-3407, pp. 337-339.*

3. Relative Contributions by Ethanol Removal Routes

Considering the effective removal flow rates for the TCC and humidity control equipment, the removal rate at a range of ethanol cabin concentrations is determined. Table 3 summarizes removal rates for each device at cabin concentrations ranging between 3.5 mg/m^3 and 25 mg/m^3. The relative removal rates in Table 3 indicate the TCCS removes 16.5% of the ethanol load and the BMP removes 62.9% of the ethanol load while the CCAA and SKV remove 9.8% and 10.9% of the load, respectively. When the TCCS has a fresh activated carbon bed installed, the removal contribution shifts with the TCCS providing 39.7% and the BMP 45.4% while the CCAA and SKV remove 7.1% and 7.8% of the load, respectively. The informative point to note is that removal via absorption by humidity condensate is small compared to the removal capability provided by the TCCS and BMP.

Table 3. Removal and total generation at varying cabin concentration.

Cabin Concentration (mg/m^3)	Scrubbing Rates (g/day)				Total Generation (g/day)	Generation above Basic* (g/day)
	TCCS	BMP	CCAA	SKV		
3	0.33	1.26	0.20	0.22	2.00	1.02
4	0.44	1.68	0.26	0.29	2.67	1.69
5	0.55	2.10	0.32	0.36	3.34	2.36
6	0.66	2.52	0.39	0.44	4.01	3.03
7	0.77	2.94	0.45	0.51	4.68	3.70
8	0.88	3.36	0.52	0.58	5.35	4.37
10	1.10	4.20	0.65	0.73	6.68	5.70
15	1.66	6.30	0.97	1.09	10.02	9.04
18	1.99	7.56	1.17	1.31	12.03	11.05
20	2.21	8.40	1.30	1.45	13.36	12.38
25	2.76	10.50	1.62	1.81	16.71	15.73

*Total generation minus 0.98 g/day.

Table 3 also highlights the total generation and the generation above the basic 0.98 g/day produced by equipment offgassing and human metabolism. The quantity above the basic rate is attributed to payload operations and crew activities and is the more variable component of the total generation. On examining Table 4, when the cabin concentration is maintained near 3 mg/m^3 as it was during the period between March 2013 and March 2014 the generation above the basic level is a little more than 1 g/day. For the 98% confidence interval range the predicted generation above the basic level ranges between 0.96 g/day and 1.39 g/day. It is most desirable to limit the generation above the basic level to close to 1 g/day to minimize water processing system impacts. These impacts are discussed in the following section.

III. Ethanol's Impact on Water Processing

The ISS Water Processor Assembly (WPA) produces potable water from a mixture of humidity condensate from the CCAA, urine distillate from the Urine Processor Assembly (UPA) and Sabatier product water. The WPA has two primary treatment steps, including the Multifiltration Beds and Catalytic Reactor. The Multifiltration Beds remove the majority of dissolved organic and inorganic contaminants via adsorbent and ion exchange media. Volatile organics not effectively removed by the adsorbents are removed downstream by the Catalytic Reactor, which oxidizes organics typically to carbon dioxide but also the corresponding acid. However, the Catalytic Reactor has a limited capacity for volatile organics. To keep the concentration of volatile organics in the condensate within the reactor capacity, the ISS Materials & Processes (M&P) organization monitors the introduction of volatile organics to the ISS environment via the Volatile Usage Agreement (VUA) process. Uses of ethanol, methanol, 1-propanol, 2-propanol, acetone, propylene glycol, and ethylene glycol aboard ISS—including payloads, systems, and flight crew equipment—are required to be reported on a VUA to insure they provide adequate containment or will release less than a total of 10 mg/day. If a known use can exceed 10 mg/day, that release is controlled by ISS operations personnel to insure the total release of volatile organics does not exceed 1 gm/day. Above this level, there is a credible risk of exceeding the capacity of the Catalytic Reactor.

In the event the reactor capacity is exceeded, ground tests have shown that competitive effects within the reactor will result in the partial oxidation of ethanol to acetic acid instead of complete oxidation to carbon dioxide. This fact is used in the concept of the Reactor Health Sensor (RHS), which detects acetic acid via an online conductivity measurement. Ground tests were used to identify the reactor's effluent conductivity that correlated to sufficient non-ionic organics (typically acetone) that would result in a violation of the potable water Total Organic Carbon (TOC) requirement. If the conductivity reaches this setpoint, the WPA is automatically transitioned to reprocess mode until the conductivity returns to acceptable levels.

Prior to 2015, there were periodic increases in the RHS conductivity indicative of increasing ethanol levels in the humidity condensate. However, these excursions typically lasted for just a few process cycles before returning to nominal levels, and were never maintained above 30 μmhos/cm. However, in early 2015, the RHS conductivity increased above the RHS setpoint of 50 μmhos/cm, requiring WPA to transition to an extended reprocess mode. Figure 2 shows the correlation between RHS conductivity and ethanol levels as reported by the Air Quality Monitor (AQM) during the first six months of 2015. Though the RHS conductivity appeared to recover when the ethanol levels were at a peak in the April/May timeframe, the low RHS value was actually because WPA only processed urine distillate during this time to recover nominal WPA operation. When the WPA began processing condensate again in June 2015, the trend again showed the correlation when the RHS conductivity decreased as the ethanol levels returned to a nominal value.

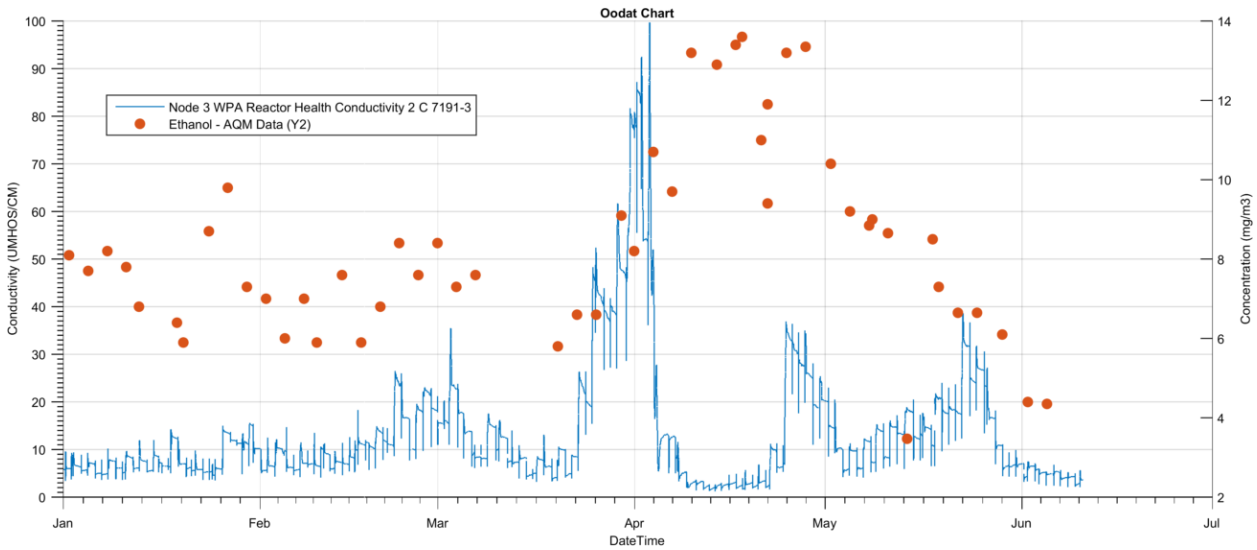

Figure 2. Influence of ethanol on the WPA Reactor Health Sensor.

A more significant concern with elevated ethanol concentration is the competitive effects with the oxidation of dimethylsilanediol (DMSD) in the reactor. DMSD was detected in the WPA product water in 2010, and now requires annual replacement of the Multifiltration Beds to maintain potable water quality. A more detailed review of DMSD and its impact on the WPA may be found elsewhere[11]. DMSD is already known to exceed the reactor capacity, since it is not completely oxidized by the catalytic reactor. The presence of elevated ethanol levels creates even more competitive oxidation effects, of which the primary concern is more unreacted DMSD being passed downstream to the Ion Exchange Bed. A higher concentration of DMSD in the influent to the Ion Exchange Bed will saturate this bed at an elevated rate, driving up the TOC in the product water and therefore requiring earlier replacement of the Multifiltration Beds. As such, it is critical to manage the ethanol at nominal levels to mitigate this impact to the TOC trend and the early replacement of the Multifiltration Beds.

IV. Ethanol Cabin Concentration Trends

Historically, NASA measured the concentrations of trace VOCs, including ethanol, using archival grab sample containers (GSCs and mini-GSCs). These samples were returned to the ground for analysis by gas chromatography/mass spectrometry using modified Environmental Protection Agency protocols[12] in the Toxicology and Environmental Chemistry (TEC) laboratory at Johnson Space Center. The mini-GSC, Fig. 3A, right, with its smaller footprint, replaced the larger GSC, Fig. 3A, left, when Shuttle flights were discontinued. Starting in April

2013, in-flight capabilities to measure target compounds, including ethanol, was established using the AQM. While the archival samples were collected once per month, it was decided that the AQM, Fig. 3B, would take measurements every 73 hours. This time interval was selected based on a balance between the volume of generated data and the stability of the ISS atmosphere, since the AQM could theoretically perform a run every 2-3 hours.

The AQM, which can be commanded to run from the ground as needed, consists of an inlet for concentrating the VOCs, a gas chromatograph (GC) to separate the VOCs, and a differential mobility spectrometer for detection of VOCs. The AQM automatically analyzes the data and stores the results to the unit and the ISS server for downlink. The crew can also view a display of results using the AQM Graphical User Interface (GUI). A more detailed technical description of the AQM operation and scientific principles can be found elsewhere.[13, 14]

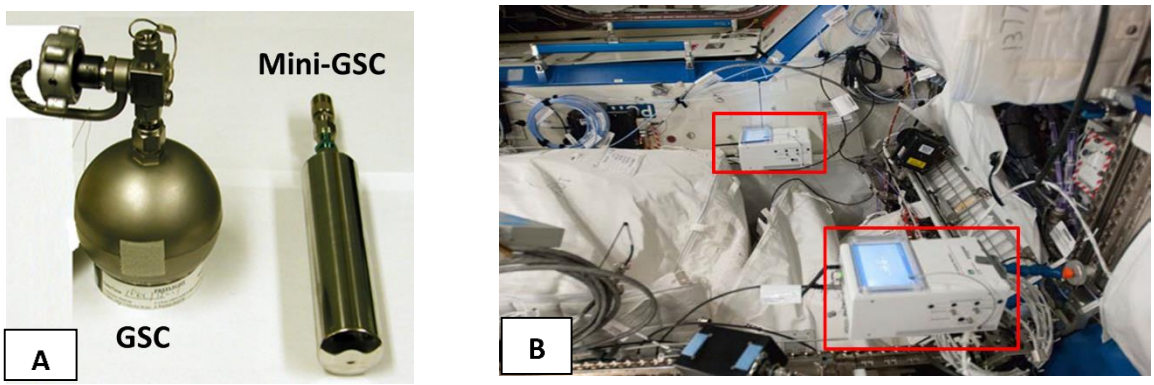

Figure 3. Air quality monitoring hardware. *A) GSC and mini-GSC, B) AQMs in ISS lab module.*

The concentrations of ethanol measured with GSCs and the AQM for October 2008 through December 2014 are shown in Fig. 4. The concentration of ethanol generally ranged from 3-6 mg/m^3 from the initiation of 6-crew operations in 2009 through early spring of 2014. Starting in early April 2014, the concentration of ethanol was extremely variable and reached a maximum of approximately 16 mg/m^3 (GSC result). When the ethanol levels are within the AQM's calibration range, the GSC results and the AQM results match quite well. When the concentrations were above the highest calibration point on the AQM, (~7.5 mg/m^3) the difference between the two methods increases. The concentration of the highest calibration point on the AQM was originally selected because atmospheric ethanol levels had been above 7 mg/m^3 in the past. The calibration range for the second set of AQMs (deployed in February 2016) was increased to ~15 mg/m^3 based on the recent trends. Although there were spikes in the ethanol concentration in July and September, they were short-lived and the level was predominantly in the 6-7 mg/m^3 range, which appeared to be the new "baseline" for 6-crew operations.

The ethanol trend for 2015 is shown in Fig. 5. A significant change in the ethanol concentration is evident, especially in April. In January and February, the ethanol concentrations were generally 6-8 mg/m^3 with one brief excursion to 10 mg/m^3. These levels are slightly above the new baseline of 6-7 mg/m^3. In late March there was a large sustained increase in reported ethanol levels that did not begin to decrease until early May. The installation of activated charcoal filters in Node 1 (intended to reduce siloxanes concentration) had only a transient effect on ethanol. There was another small increase in July followed by a much larger, prolonged ethanol increase in mid-August before the concentration stabilized at a lower concentration in late September.

The GSC and AQM values showed excellent agreement through May 2015. However, from June through November there was a ~45% difference between the AQM and GSC results, though the trend in the ethanol concentration was the same for both techniques. The GSC values were higher even when the concentrations were within the AQM calibration range. This was attributed to a decrease in the sensitivity of the AQM as it approached the end of its operational lifetime. Although the AQM's October through December values are ~3-4 mg/m^3, the reduced sensitivity for this unit and the GSC results suggest the ethanol concentrations are approximately at the new baseline (~6-7 mg/m^3). Both represented a substantial decrease in the ethanol concentration from the August timeframe.

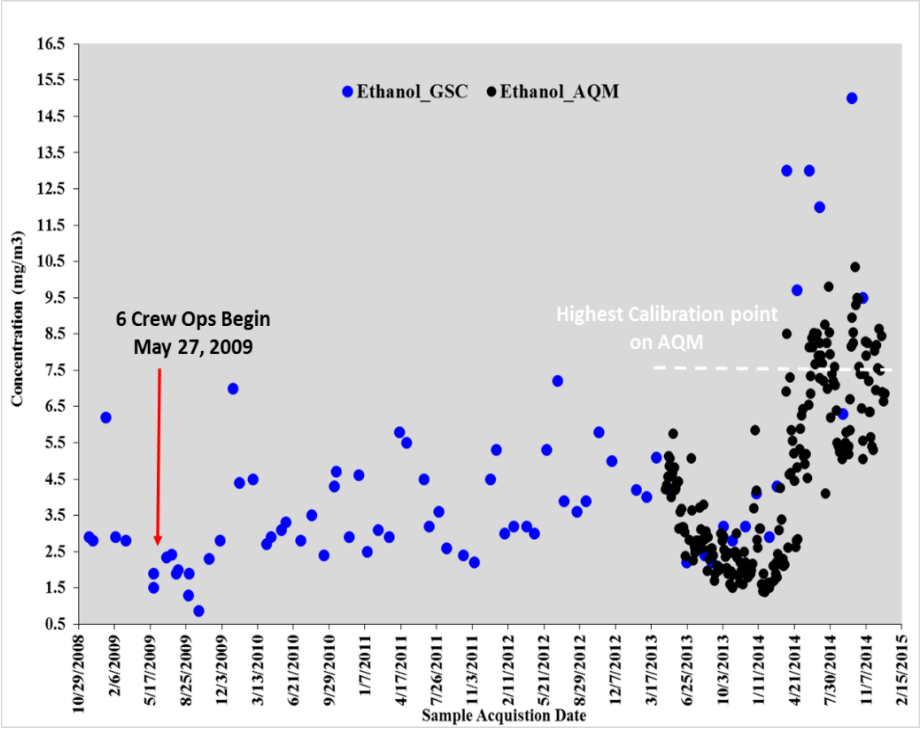

Figure 4. GSC and AQM ethanol data from October 2008 through December 2014. *Only GSCs collected in the lab module are shown. Data for the AQM is shown from the beginning of operations in April 2013 through December 2014.*

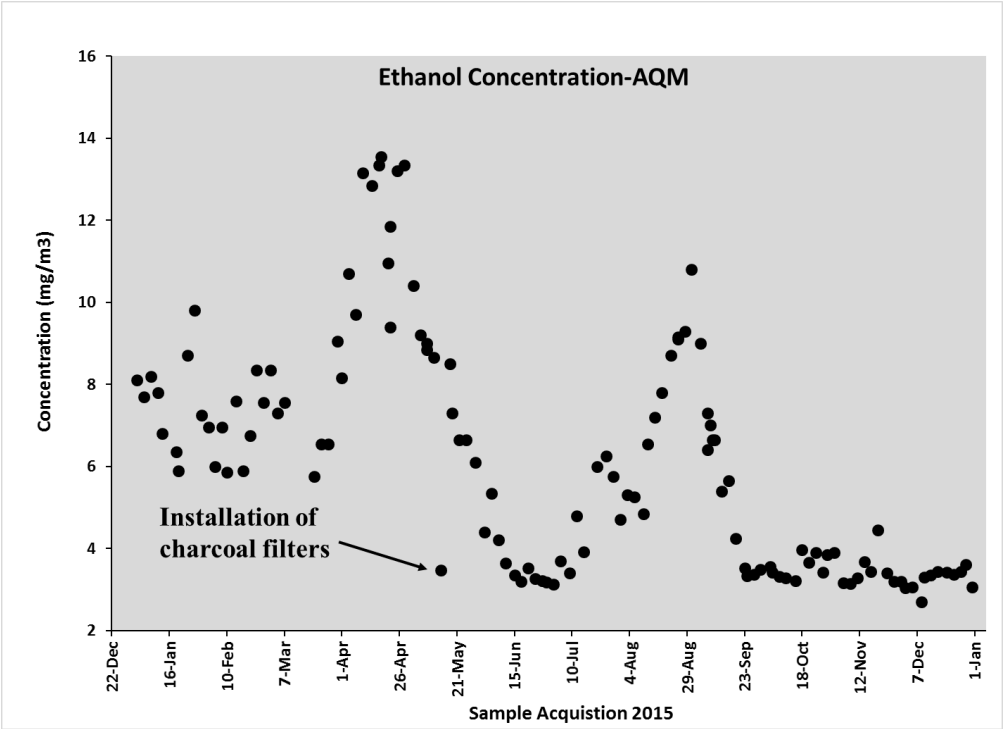

Figure 5. AQM ethanol data for January 2015 through December 2015. *Two concentration increases were observed during this period.*

The WPA conductivity began to increase during April 2015, which coincided with the sustained increase in atmospheric ethanol concentrations. This connection between the WPA conductivity and the atmospheric ethanol concentration launched the investigation into potential sources of ethanol on ISS. The plots of WPA conductivity and ethanol concentration are shown in Fig. 6. The trending shows a definite correlation between the WPA RHS conductivity and AQM readings, especially when taking into account the vastly different sampling times for the two parameters and the periods when condensate was not being processed.

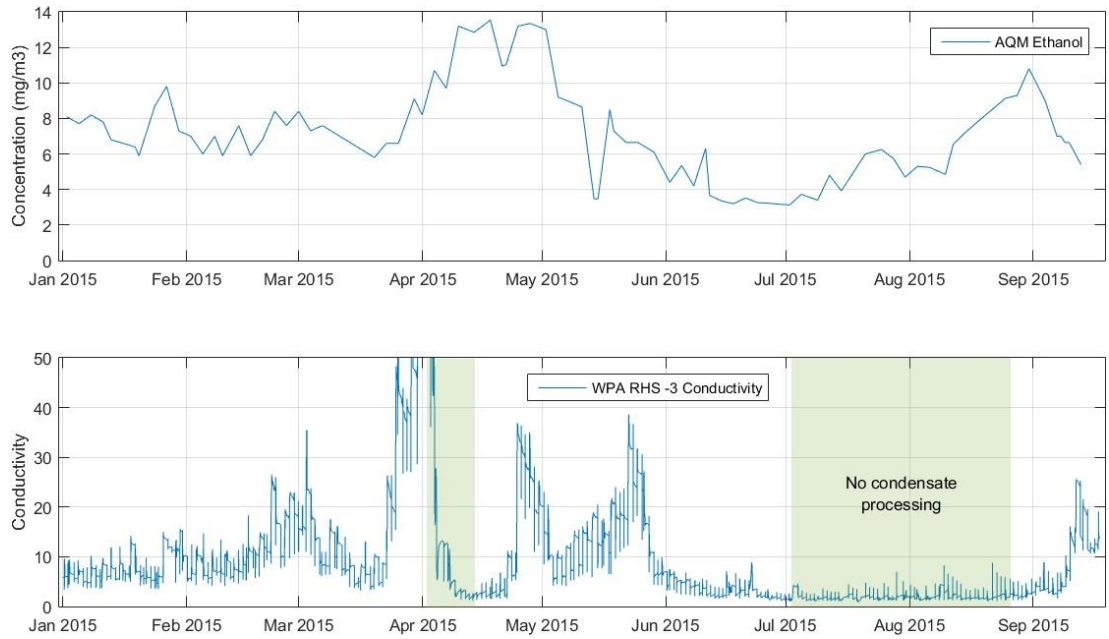

Figure 6. Ethanol concentration compared to WPA conductivity. *The upper graph trends the ethanol concentration as reported by the AQM and the lower graph shows the WPA conductivity over the same time period.*

V. Potential Ethanol Sources Contributing to Concentration Increases

After the elevated ethanol concentration was observed on ISS, a multidisciplinary team was formed to review credible sources of ethanol on ISS and develop recommendations to reduce ethanol levels in the atmosphere. This review was conducted as part of a Multilateral Anomaly Resolution Team (MART) to insure input from the international partners that contribute payloads, crew, and systems to the ISS environment. The VUA process managed by ISS M&P was also reviewed in detail to verify the acceptability of this process and identify any potential paths that could circumvent this process and allow an excessive quantity of ethanol on ISS. While many theories were evaluated for merit, the AQM results and source generation analysis determined that a credible source must not be transient in nature, but must be relatively significant and sustained to explain the levels observed on ISS.

Ground personnel reviewed crew hygiene items, housekeeping items, payloads, ISS systems and ground processing of vehicles prior to launch for potential sources of ethanol. The most significant source of ethanol that could be identified is the Veltosept wipes used in the Russian Segment for housekeeping and payloads use, but the total quantity of ethanol in these wipes (<100 g/year) is significantly less than that required to produce the ethanol levels observed in the ISS atmosphere (>10 g/day per Table 3). Ethanol wipes are also used for housekeeping purposes in the U.S. Segment, but these wipes contain less ethanol than the Veltosept wipes, and are used at a lower frequency. The only payloads that might use ethanol included the Rodent Research Facility which does not appear to be using any ethanol in their operations at this time.

Another credible source is crew hygiene items, such as perfumes, colognes, and deodorant. JAXA's skin cleaner was evaluated and found to contain a relatively low quantity of ethanol compared to the other credible sources. Overall, the nominal use of crew hygiene items was found to produce ethanol at rates measurably below the generation rate required to produce the ethanol levels observed on ISS.

Finally, concentration spikes are observed during vehicle docking operations and some crew activities; however, these excursions are not sustained like those observed in this instance. Again, in these limited instances, the quantity

of ethanol released into the ISS atmosphere is neither sufficient nor sustained to result in the observed concentration increase.

In summary, after an extensive review of credible sources on ISS, no significant contributor could be identified.

VI. Conclusion

A significant increase in atmospheric ethanol concentration was observed on ISS in 2015, culminating in a general increase in ethanol on ISS since initially crewed. Though elevated ethanol concentrations are not a concern for crew health, they can impact the performance of the WPA by exceeding the reactor oxidation capacity and impacting the life of the Multifiltration Beds due to competitive effects in the reactor with DMSD. After a thorough review of credible sources on ISS, no significant contributor could be identified. Further, ISS crew were questioned on possible activities that could contribute to the elevated ethanol levels, but no likely source was identified. Fortunately, in June 2015 the ethanol levels returned to nominal concentrations. Though obvious excursions have been detected by the AQM since then, no sustained increase in ethanol has been maintained. Ground personnel will continue to monitor this trend and discuss credible sources with the ISS crew in an attempt to resolve the source and mitigate future excursions.

Acknowledgments

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