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PURPOSE

The purpose of this report is to summarize the analyses performed under the ORD-FY04-017 “Yucca Mountain Saturated Zone Carbon-14” DOE project. The work was performed under Cooperative Agreement No. DE-FC28-04RW12232 between the U.S. Department of Energy and the Nevada System of Higher Education (NSHE). The research was conducted by collecting groundwater samples in the Yucca Mountain area, processing samples at the Desert Research Institute, and having carbon dioxide gas from the processed samples analyzed at the University of Arizona Accelerator Facility from May 2004 to December 2006. This task included developing implementing procedures for evaporating dissolved organic carbon water samples (IPR-029; see Appendix A for procedure description) and combusting dissolved organic carbon samples (IPR-030; see Appendix B for procedure description).

This report presents the dissolved organic carbon isotopic data collected, processed, and analyzed for this study. Geochemical modeling using the computer program NETPATH (a NSHE QA’ed approved model program) was also conducted as part of this study to determine groundwater sources, flow paths, mixing, and ages (travel times) for water in the Yucca Mountain area. The geochemical modeling was conducted using major ion chemistry, deuterium and oxygen-18, and dissolved inorganic carbon (DIC) isotopic data from the Yucca Mountain QA data base. However, although the majority of work on the modeling task was completed, because full funding was not provided for the project, final technical and QA reviews of the resulting data and analyses could not be completed. Therefore the results of the groundwater geochemical modeling and associated travel times as well as a comparison of DIC and DOC groundwater travel times in the Yucca Mountain area to each other and to groundwater travel times developed from groundwater flow models are not included in this report. The information has, however, been submitted to the Yucca Mountain Records Information System. Finally, because the project was not fully funded this is not a QA report.

QUALITY ASSURANCE

All data generated by this project were produced in accordance with the NSHE Quality Assurance Program. However, since funding was terminated prematurely, final technical and QA reviews could not be completed. Therefore the results presented in this report are unqualified and may not be used for quality affecting work.

INTRODUCTION

Groundwater samples in the Yucca Mountain area were collected for chemical and isotopic analyses and measurements of water temperature, pH, specific conductivity, and alkalinity were obtained at the well or spring at the time of sampling. For this project, groundwater samples were analyzed for major-ion chemistry, deuterium, oxygen-18, and carbon isotopes of dissolved inorganic carbon (DIC) and dissolved organic carbon (DOC). The U.S. Geological Survey (USGS) performed all the fieldwork on this project including measurement of water chemistry field parameters and sample collection. The major ions dissolved in the groundwater, deuterium, oxygen-18, and carbon isotopes of dissolved inorganic carbon (DIC) were analyzed by the USGS. All preparation and processing of samples for DOC carbon isotopic analyses and geochemical modeling were performed by the Desert Research Institute (DRI). Analysis of the DOC carbon dioxide gas produced at DRI to obtain carbon-13 and carbon-14 values was conducted at the University of Arizona Accelerator Facility (a NSHE Yucca Mountain project QA qualified contract facility). The major-ion chemistry, deuterium, oxygen-18, and carbon isotopes of DIC were used in geochemical modeling (NETPATH) to determine groundwater sources, flow paths, mixing, and ages. The carbon isotopes of DOC were used to calculate groundwater ages that are independent of DIC model corrected carbon-14 ages. The DIC model corrected carbon-14 calculated ages were used to evaluate groundwater travel times for mixtures of water including water beneath Yucca Mountain. When possible, groundwater travel times were calculated for groundwater flow from beneath Yucca Mountain to down gradient sample sites. DOC carbon-14 groundwater ages were also calculated for groundwaters in the Yucca Mountain area. When possible, groundwater travel times were estimated for groundwater flow from beneath Yucca Mountain to down gradient groundwater sample sites using the DOC calculated groundwater ages. The DIC calculated groundwater ages were compared with DOC calculated groundwater ages and both of these ages were compared to travel times developed in ground-water flow and transport models.

If nuclear waste is stored in Yucca Mountain, the saturated zone is the final barrier against the release of radionuclides to the environment. The most recent rendition of the TSPA takes little credit for the presence of the saturated zone and is a testament to the inadequate understanding of this important barrier. If radionuclides reach the saturated zone beneath Yucca Mountain, then there is a travel time before they would leave the Yucca Mountain area and flow down gradient to the Amargosa Valley area. Knowing how long it takes groundwater in the saturated zone to flow from beneath Yucca Mountain to down gradient areas is critical information for potential radionuclide transport. Radionuclide transport in groundwater may be the quickest pathway for radionuclides in the proposed Yucca Mountain repository to reach land surface by way of groundwater pumped in Amargosa Valley. An alternative approach to ground-water flow and transport models to determine the travel time of radionuclides from beneath Yucca Mountain to down gradient areas in the saturated zone is by carbon-14 dating of both inorganic and organic carbon dissolved in the groundwater. A standard method of determining ground-water ages is to measure the carbon-13 and carbon-14 of DIC in the groundwater and then correct the measured carbon-14 along a flow path for geochemical reactions that involve carbon containing phases. These geochemical reactions are constrained by carbon-13 and isotopic fractionations. Without correcting for geochemical reactions, the ground-water ages calculated from only the differences in carbon-14 measured along a flow path (assuming the decrease in carbon-14 is due strictly to radioactive decay) could be tens of thousands of years too old. The computer program NETPATH, developed by the USGS, is the best geochemical program for correcting carbon-14 activities for geochemical reactions. The DIC carbon-14 corrected ages can be further constrained by measuring the carbon isotopes of DOC. Because the only source of organic carbon in aquifers is almost always greater than 40,000 years old, any organic carbon that may be added to the groundwater would contain no carbon-14. Thus, ground-water ages determined by carbon isotopes of DOC should be maximum ages that can be used to constrain DIC corrected ages.

METHODS AND MATERIALS

All field work was performed by the U.S. Geological Survey (USGS) and all laboratory work involving DOC samples was performed by the Desert Research Institute (DRI) and the University of Arizona. All DOC samples collected by the USGS were transported to DRI and from DRI to the University of Arizona using NSHE chain of custody procedures. Sample processing at DRI was performed using NSHE approved implementing procedures IPR-029 (Appendix A; evaporating dissolved organic carbon water samples) and IPR-030 (Appendix B; combusting dissolved organic carbon samples). All geochemical modeling was done using the NSHE Yucca Mountain QA/QC approved NETPATH computer program with data from Yucca Mountain qualified data base. All samples collected and analyzed for this project were collected and analyzed using Yucca Mountain QA/QC approved methods.

The USGS computer program NETPATH was used to model the changes in groundwater chemistry along potential groundwater flow paths to evaluate the sources and mixing of different waters in the Yucca Mountain area. In order for a potential flow path to be a valid flow path, or for a valid mixture of more than one groundwater, the observed changes in groundwater chemistry have to be reasonable with known mineral, salt, or gas dissolution and thermodynamically feasible (minerals the dissolve need to be under saturated in the water and minerals that form need to be over saturated in the water). The geochemical models were also constrained by deuterium. The down gradient groundwater for all flow paths and mixtures of groundwaters had to be within 3 permil of measured values for the flow path and/or mixing geochemical model to be acceptable.

ASSUMPTIONS

No assumptions were made in conducting this work.

DISCUSSION AND CONCLUSIONS

Sites of groundwater samples for which data has been entered into the NSHE qualified data base in this report are shown in Figure 1. The UTM and latitude/longitude locations of the sample sites are given in Table 1. Results of sample analyses are presented in Table 2. These data have been entered into the NSHE Technical Data Archive and have been incorporated into the Yucca Mountain Technical Data Managament System.

Table 1: Location of DOC groundwater sample sites with data entered into the NSHE Technical Data Archive (see Figure 1 for map with samples located on them). SAMPLE_NAME is the well or spring name; UTM_X and UTM_Y are the UTM locations; and LAT_DMS and LONG_DMS are latitude and longitude in degrees, minutes, and seconds.

LOCAL_NAME	UTM_X	UTM_Y	LAT_DMS	LONG_DMS
Ash Meadows	559643.3	4030384	362507	1162005
Gilgans #1	549744.6	4036731	362835	1162641
MSH-C Shallow well	565375.6	4039701	363008	1161612
MSH-C Deep well	565375.6	4039701	363008	1161612
NDOT-2	553685.4	4055242	363835	1162358
USW VH-2	537738.3	4073214	364821	1163437
NC-EWDP-01DX	536847.8	4062508	364234	1163515
NC-EWDP-10P Zone 1	553149.0	4064916	364349	1162417
NC-EWDP-12PA	536984.8	4060772	364137	1163510
NC-EWDP-12PB	536951.9	4060799	364138	1163511
NC-EWDP-15P	544926.9	4058163	364011	1162950
NC-EWDP-16P	545665.0	4064263	364329	1162919
NC-EWDP-18P	549415.5	4067233	364505	1162647
NC-EWDP-19PB Zone 1	549336.7	4058316	364015	1164479
NC-EWDP-19PB Zone 2	549336.7	4058316	364015	1164479
NC-EWDP-22PA Zone 1	552020.1	4062038	364215	1162503
NC-EWDP-22PA Zone 2	552020.1	4062038	364215	1162503
NC-EWDP-24P	549375.0	4062026	364216	1162650
NC-EWDP-27P	544935.2	4065275	364402	1162948
NC-EWDP-29P	549396.5	4059607	364057	1162650

Table 2: DOC groundwater sample analyses results. Sample Name is the well or spring name; NA, data are not available (DID: 017JT.001, DTN: MO0705UCC017JT.001).

Sample Name	Sample Date	DOC (mg/l)	Carbon in gas (mg)	Carbon after gas cleanup (mg/l)	^{13}C (‰)	^{14}C (pmc)	^{14}C error (pmc)
Ash Meadows	7/12/2005	NA	0.12	0.12	-34.6	30.2	1.3
Ash Meadows	7/12/2005	0.34	0.10	0.10	-30.5	34.3	0.6
Gilgans #1	7/13/2005	NA	0.14	0.13	-34.2	31.1	0.6
MSHC-Shallow	7/12/2005	NA	0.09	0.09	-31.2	22.7	1.0
MSHC-Deep	7/12/2005	3.46	0.22	0.22	-31.7	33.8	0.4
NDOT 2	7/12/2005	NA	0.58	0.44	-38.3	25.1	0.2
USW VH2	3/1/2001	NA	0.13	0.13	-36.4	0.7	0.7
NC-EWDP-01DX	9/29/2005	0.49	0.53	0.53	-30.8	14.4	0.2
NC-EWDP-10P Zone1	8/27/2002	0.21	0.17	0.17	-35.9	41.2	0.6
NC-EWDP-12PA	9/27/2005	0.28	0.26	0.26	-35.4	17.6	0.4
NC-EWDP-12PB	9/28/2005	0.51	0.20	0.19	-32.7	25.1	0.5
NC-EWDP-15P	10/26/2000	NA	0.11	0.11	-31.6	85.2	0.9
NC-EWDP-16P	9/20/2005	1.18	0.05	0.05	-28.8	36.7	1.4
NC-EWDP-18P	8/26/2002	0.96	0.85	0.85	-31.3	41.2	0.3
NC-EWDP-19PB Zone 1	5/11/2004	0.24	0.13	0.09	-36.7	42.9	0.7
NC-EWDP-19PB Zone 1	5/11/2004	0.24	0.09	0.09	-26.3	44.3	0.9
NC-EWDP-19PB Zone2	5/10/2004	0.24	0.11	NA	-32.9	31.6	0.3
NC-EWDP-22PA Zone 1	8/28/2002	0.16	0.27	NA	-27.3	32.1	0.4
NC-EWDP-22PA Zone1	8/28/2002	0.16	0.07	0.07	-31.2	35.7	1.2
NC-EWDP--22PA Zone2	8/28/2002	0.17	0.06	0.06	-26.3	51.1	1.2
NC-EWDP-24P	9/22/2005	0.24	0.40	0.38	-35.6	20.7	0.3
NC-EWDP-27P	9/19/2005	0.42	0.18	0.18	-32.4	22.4	0.6
NC-EWDP-29P	9/23/2005	0.35	0.11	0.11	-39.2	23.6	1.2

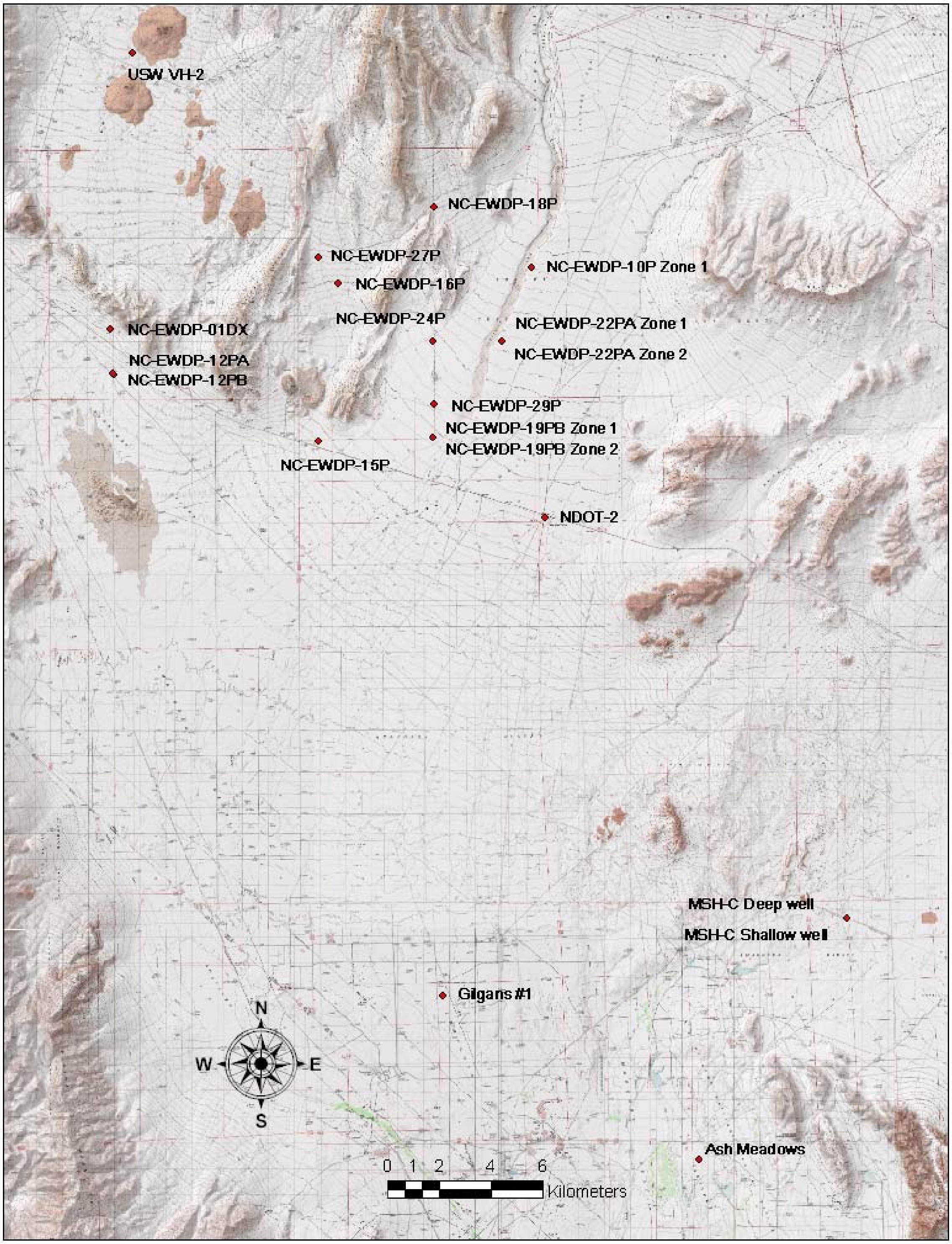

Figure 1: Map showing sample site locations.

APPENDIX A. Implementing Procedure for Evaporating Dissolved Organic Carbon Samples (IPR-029)

Title: Evaporating Dissolved Organic Carbon Samples
Procedure No.: IPR-029, Rev. 0

1.0 PURPOSE AND SCOPE

The purpose of this implementing procedure (IP) is to ensure that collection (described in detail in scientific notebook) and preparation of dissolved organic carbon (DOC) samples from Yucca Mountain sites are properly conducted prior to and during evaporation and that they meet quality assurance standards of the UCCSN QA Program. This IP describes the evaporation method that will be applied to DOC samples and how maintaining certain temperature and pressure readings will contribute to the objective of a standardized method of evaporation. Sample preparation, not data acquisition, is the key objective, in this procedure. DOC samples are collected and labeled using site names and coordinates for identification purposes in accordance with QAP 8.0, Identification and Control of Samples. The evaporation equipment is located in the Organic Carbon Laboratory of the Maxey Building of The Desert Research Institute, Reno Nevada.

2.0 APPLICABILITY

This procedure applies to Yucca Mountain DOC samples prior to combustion and to the personnel assigned to the evaporation process.

3.0 PROCEDURE

3.1 Any work conducted in accordance with this IP must abide to the parameters and instructions. When unable to comply with the specified steps, the work must be suspended and resumed only after the procedure has been revised or a Document Change Notice has been documented. There are no special standards or criteria for this work and no specific job skill other than those stated in the position descriptions filed with HRC.

3.1.1 Samples are identified by site name/location and are custom sealed at collection, opened just prior to evaporation. In accordance with QAP 8.0, the samples are collected in 1L oven fired, brown sample bottles, which are clearly labeled with the site name and location coordinates and time/date of collection and collector's initials and sealed with custom seals bearing the collector's initials and date/ time of collection. The DOC and samples are then transported to the laboratory in ice packed coolers and immediately refrigerated in designated refrigerators according to site name and date of collection. Refrigerated samples have a shelf life of about two years. Any alteration in the control and handling of the samples and glitches in the readings would be noted in Scientific Notebook for future referencing and troubleshooting to ensure reproducibility.

3.2 Sample Preparation

1. Document the details of sample collection in the scientific notebook.
2. Transfer sample from collection container to sterilized (heated in furnace at 800 C for 15 minutes) quartz bottle.
3. Add 20 drops of hydrochloric acid to sample.

4. Use pH paper to ensure that sample reads pH <3.
5. Attach sample bottle to evaporation equipment with plastic tubing and fittings.

3.3 Evaporation Equipment Operation--Turn on Procedure

1. Fill inner container to the rim with alcohol 90% ethanol-Ethyl Alcohol UN1170.
2. Reassemble cold trap (wider outer ring first, then inner container, then cold trap arm, then foil insulator disc, then the lid since the arm is cold and brittle, hence it can break easily).
3. Check that temperature of cold trap alcohol is less than -10$_o$C (merely a guideline, doesn't need to be a specific temperature < -10$_o$C.
4. Check that the red valve is parallel to line.
5. Check that the vacuum pump is working by turning on pump (plug into socket). Below 10mbar is good, 5 to 6 mbar better.
6. Check that the green valves closed (perpendicular to line).
7. Open cold trap valve (red), then open valve to vacuum pump and wait for pressure to read < 10 mbar.
8. Tuck temperature controller probe between bottle and heating jacket. Insulate from above with sponge ring.
9. Plug temperature controller into socket and follow directions on pamphlet for heater mode (Use 28$_o$C standard). Finally open valve for sample (green valve closest to bottle).
10. Record vacuum pump reading, cold trap temperature, estimated volume of sample (required for the first evaporation only) and start time in Scientific Notebook.

3.4 Evaporation Equipment Operation--Turn off Procedure

1. Turn off valve for sample (closest to Bottle).
2. Turn off valve for vacuum pump (closest to the gauge).
3. Turn off heating jacket (unplug).
4. Turn off vacuum pump (unplug).
5. Turn off Neslab.
6. Release red valve at a diagonal.
7. Remove cold trap arm and place in beaker (let thaw out for 2hrs to prevent breakage).
8. Remove cold trap inner container (keep lid on to retain alcohol) and remove ice from sides by running under hot water. Thoroughly dry outside surface of inner container and interior of outer container.

3.5 Maintain Scientific Notebook

Keep Scientific Notebook locked in fire-rated cabinet in locked laboratory located in building accessed only by authorized pass keys until completion of project or full use of scientific notebook, whereby Principal Investigator assumes custody.

4.0 Quality Assurance Records

Quality Assurance Records that result from the implementation of this procedure are controlled and transmitted to the Document Control Coordinator in Accordance with QAP-17.0, “Quality Assurance Records,” and include None.

APPENDIX B. Implementing Procedure for Combusting Dissolved Organic Carbon Samples (IPR-030)

Title: Combusting Dissolved Organic Carbon Samples
Procedure No.: IPR-030, Rev. 0

1.0 PURPOSE AND SCOPE

The purpose of this implementing procedure is to combust dissolved organic carbon (DOC) Yucca Mountain samples using a custom made combustion line and to ensure that the handling of the previously evaporated DOC samples meet the quality assurance standards of the UCCSN QA Program.

This IP describes the combustion method that will be applied to DOC samples and how maintaining certain temperature and pressure readings will contribute to the objective of a standardized method of combustion. Actual preparation of the sample and its combustion is the key objective, not the acquisition of data. The combustion line is located in the Organic Carbon Laboratory of the Maxey Building of The Desert Research Institute, Reno Nevada.

2.0 APPLICABILITY

This procedure applies to previously evaporated Yucca Mountain DOC samples and to the personnel assigned to the combustion process.

3.0 PROCEDURE

Any work conducted in accordance with this IP must abide to the parameters and instructions. When unable to comply with the specified steps, the work must be suspended and resumed only after the procedure has been revised or a Document Change Notice has been documented.

There are no special standards or criteria for this work and no specific job skill other than those stated in the position descriptions filed with HRC.

Sample Control and Handling

Samples are identified by site name/location and are custom sealed at collection, opened just prior to evaporation and combusted directly thereafter. The sample vessel (labeled with site name, date of collection) containing the evaporated DOC sample is detached from the evaporation equipment and attached to the combustion line. Evaporated samples have a shelf life of a year and need to be kept sealed in sample vessel until combustion. Since both sets of equipment are located in the same laboratory and both processes conducted by the same laboratory technician, there is no change of custody and the procedure is in accordance with QAP 8.0, Identification and Control of Samples. Any alteration in the control and handling of samples and glitches in the readings would be noted in the Scientific Notebook for future referencing and troubleshooting to ensure reproducibility.

1. Place a sample collection vessel and an evaporated water sample on the line.
2. Insert the water thermocouple between the furnace and the water sample vessel. (This is done at this time as a safety precaution.)

3. Switch the vacuum/scroll pump to roughing and rough pump the line up to the blue valve. Then rough pump the line up to and including the sample vessel.
    a. For rough pump operation: i) close the butterfly valve at the top of the turbo pump (groove in butterfly valve knob needs to be horizontal), and ii) switch the bypass valve from backing to roughing. Then starting from the biggest valve (above right of the turbo pump) open valves in sequence from pump up to but not including blue valve just before sample. [Exclude the red valve directly opposite blue valve and the valve diagonally across from the red valve since this is an alternative flow path]. Once gauge reading drops to 10^{-2} torr then open blue valve.
    b. Once gauge reading drops again to 10^{-2} torr then open valve to sample vessel. [Note; if gauge reading doesn't reach 10^{-2} torr after blue valve has been opened about 1hr then disconnect quartz tubing and reconnect to ensure a proper vacuum is obtained, which will be indicated by 10^{-2} torr].
4. When the vacuum falls below 10^{-2} torr, switch to the turbo pump. For turbo pump operation: i) switch the bypass valve from roughing to backing, and ii) open the butterfly valve at the top of the turbo pump (groove in butterfly valve needs to be vertical). Plug in the turbo pump.
5. Let the water sample pump to less than 10^{-3} torr.
6. Prepare three dewars: i) short one with an ethanol/liquid nitrogen slurry* and ii) two tall dewars with liquid nitrogen. Put one of the liquid nitrogen dewars on the turbo pump cold finger.

    * To prepare the slurry, slowly add LN (liquid nitrogen) to the alcohol and stir gently with a glass rod or tube. The slurry will become thicker and eventually solid ethanol will form on the surface of the liquid. Carefully knock this "ice" down into the dewar. The slurry should be ready at this point. Check the temperature with the thermocouple; it should be around -115 °C. If it is not cool enough keep adding LN in small increments and stirring into solution until the desired temperature is reached. Carefully place the alcohol slurry on the spiral trap.
7. Place a tall dewar with liquid nitrogen on the first cold finger beyond the spiral trap (sample cold finger).
8. Turn on platinum furnace.
9. Add oxygen gas to sample. Check to see that the supply valve at the oxygen tank is open. The regulator should be set to 20 psi. Close the sample to the turbo pump (i.e., close the red valves, valve on line above green valve and, using the green oxygen supply valve, bring the pressure in the sample chamber to –25 in. Hg (measured on the mechanical combustion line gauge).
10. Close the blue valve. Close the green valve and the valve on line above the green valve. Close the valve to the oxygen line. [This is a safety precaution]. The sample is now ready to be combusted.
11. Adjust the valves on the line to make a pathway from the sample combustion section up to the bleed valve (marked with a "B"), passing through the platinum furnace. Keep the bleed valve closed. The valve above the sample cold finger and the valve to the known volume section should be open.
12. Adjust the furnace temperature controller to 500 °C.

    a. To adjust the heater, hold down the left button (which has an asterisk*) and use the up button to increase the set point or the down button to decrease the set point.
    b. Caution! If the thermocouple is not in the heater, the heater will heat to its maximum level.
13. Ignite the Bunsen burner and heat the sample vessel at 500 °C for 15 min (it will take approx. 20 min to reach the desired temperature).
14. After sample has combusted for 15 min, slowly open the line to the vacuum pump using the bleed valve while keeping the pressure below 10^{-1} torr. This step of freezing out the CO_2 is a delicate step requiring numerous adjustments of the valve. This step can take a half-hour or more to perform. Explanation: the sample gas contains water vapor, CO_2 and oxygen (among other gases). The idea is to freeze the water in the spiral trap, and to freeze the CO_2 in the sample cold finger while removing oxygen with the turbo pump. It is important to maintain low pressure ($<10^{-1}$ torr) so as not to lose any of the CO_2.
15. Watch the combustion vacuum gauge. When it falls to -30 in. Hg, open the valve above the cold finger and pump to less than 10^{-5} torr.
16. Close the blue valve. Turn off the Bunsen burner. Turn off platinum furnace.
17. Close the oxygen supply at the tank.
18. Adjust the furnace controller to less than 20 °C (This is a safety precaution!) and then shut off.
19. Adjust valves to place vacuum in known volume before transferring sample if needed (this is a safety precaution).
20. Freeze sample from the cold finger into the first finger of the known volume (~10 min). This will be indicated by a clear inner tube of the cold finger and increasing readings on the Baratron gauge.
21. With the sample frozen in the known volume, pump briefly (by opening the extreme left valve of known volume section) to remove residual O_2.
22. Isolate the known volume (close valves on all sides) and allow the gas to sublimate (remove liquid nitrogen). Watch the Baratron gauge for decreasing readings, indicating the gradual release of sample from the first finger of the known volume and its readiness to be transferred to the sample tube.
23. Place dewar with liquid nitrogen on sample tube.
24. Measure the volume of the sample and note it in the Scientific Notebook and transfer the sample into a sealed tube for shipment to Arizona.
25. Plug the measured volume value into the known volume formula noted at the beginning of the Scientific Notebook to calculate the mass of the combusted sample. Note the calculated mass in the Scientific Notebook and on the label of collection vessel to be shipped. Note also the Principal Investigator's name and date of combustion on the label.
26. Fill UCCSN Chain of Custody Form per QAP 8.2, Sample Transfer, and ship with vessel containing combusted sample.
27. Keep Scientific Notebook locked in a fire-rated cabinet in a locked laboratory located in building accessed only by authorized pass keys until completion of project or full use of the Scientific Notebook, whereby Principal Investigator assumes custody.

4.0 Quality Assurance Records

Quality Assurance Records that result from the implementation of this procedure are controlled and transmitted to the Document Control Coordinator in Accordance with QAP-17.0, “Quality Assurance Records,” and include None.