

# National Human Exposure Assessment Survey (NHEXAS)

## *Maryland Study*

### Quality Systems and Implementation Plan for Human Exposure Assessment

Emory University  
Atlanta, GA 30322

Cooperative Agreement CR 822038

**Standard Operating Procedure**

**NHX/SOP-F08**

**Title:** Collection, Storage, and Shipment of Duplicate Diet  
Samples for Metal, Pesticide, and PAH Analysis

**Source:** Harvard University/Johns Hopkins University

U.S. Environmental Protection Agency  
Office of Research and Development  
Human Exposure & Atmospheric Sciences Division  
Human Exposure Research Branch

**Notice:** The U.S. Environmental Protection Agency (EPA), through its Office of Research and Development (ORD), partially funded and collaborated in the research described here. This protocol is part of the Quality Systems Implementation Plan (QSIP) that was reviewed by the EPA and approved for use in this demonstration/scoping study. Mention of trade names or commercial products does not constitute endorsement or recommendation by EPA for use.

1 Title of Standard Operating Procedure

Harvard University/Johns Hopkins University Standard Operating Procedures:

**F08 Collection, Storage, and Shipment of Duplicate Diet Samples for Metal, Pesticide, and PAH Analysis, Rev. 1.0**

2 Overview and Purpose

The purpose of this SOP is to describe techniques for the collection and handling of duplicate diet samples in the field. The duplicate diet sample is a four-day composite sample of all foods and beverages consumed by the target individual, whether prepared or consumed in the home or elsewhere. To reduce burden, the target individual may choose which days to save food.

3 Discussion

Food may be an important pathway for exposure to environmental contaminants. In this investigation, we will gather samples of foods and beverages consumed by the target individual, and prepare them for analysis. This four-day, aggregate, duplicate diet sample will be compared to the Mini-Market Basket survey sample to determine the adequacy of the latter in determining dietary exposure to pollutants.

4 Personnel Responsibilities

4.1 Sampler Preparation

Arranging appointments, asking whether the respondent wants a refrigerator, and making reminder calls are the responsibility of the Field Coordination Center (FCC) staff.

Preparation of equipment and instruction of the respondent are the responsibility of the Field Interviewer.

4.2 Sample Collection

Field collection during the sampling period is the responsibility of the respondent. Recovery of the samples at the end of the sampling period is the responsibility of the Field Interviewer.

4.3 Storage

Storage of the samples during the four-day duplicate diet study is the responsibility of the respondent. Handling of the collected samples prior to delivery to the FCC, and aggregation and weighing of the samples at the FCC, are the responsibility of the Field Interviewer. Handling and storage at the FCC prior to shipping is the responsibility of the Field Coordinator or his designate.

#### 4.4 Shipment

Shipment to the cooperating analytical laboratory (assumed to be FDA) is the responsibility of the Field Coordinator or his designate.

#### 4.5 Analysis

Analysis of all food samples is the responsibility of the coordinating laboratory (assumed to be FDA).

### 5 Required Equipment and Reagents

#### 5.1 Field Sampling

##### 5.1.1 Day 1

refrigerator, about 2.5 cubic feet (or carton to transport containers etc.) with duplicate diet instructions taped to door  
reusable cold packs, total up to 5" x 5" x 12"  
(to be stored in freezer of small refrigerator or household refrigerator)  
12 beverage storage jars with lids: polyethylene, 2-quart, acid washed (one-time use)  
20 plastic bags, resealable (Ziplok or equivalent), 3-quart  
handcart, bungee cords, straps for moving refrigerator

##### 5.1.2 Day 8

food storage cooler, 60-80 quart (if respondent did not use refrigerator)  
handcart, bungee cords, straps for moving refrigerator

#### 5.2 Sample Tracking and Paperwork

Food Checklist and instruction sheet with telephone number  
3 sample ID labels (sample type 07, for food checklist):  
1 on checklist, 1 on chain-of-custody form, 1 spare  
Duplicate Diet instruction sheet with telephone number to give to target individual  
30 sample ID labels (sample type 51, for duplicate diet - solid foods): 20 on sample bags,  
1 on refrigerator or cooler, 1 on chain-of-custody form, 3 on logsheet, 2 for aggregate containers, 3 spare  
20 sample ID labels (sample type 52, for duplicate diet - beverages): 12 on sample jars,  
1 on refrigerator or cooler, 1 on chain-of-custody form, 3 on logsheet, 3 spare  
Field Packet for household:  
logsheet (3-part carbonless)  
chain-of-custody form for food checklist  
chain-of-custody form for duplicate diet samples  
Field Manual (SOPs)  
clipboard  
pens, ballpoint

#### 5.3 Field Coordination Center (FCC) -- after field sampling (Day 8)

rubber scraper  
containers for aggregate sample -- 4-L plastic jars  
scale for weighing aggregate sample: capacity 5 kg, precision 1 g

## 6 Procedure

### 6.1 Preparation for Collection

#### 6.1.1 Reminder to Respondent (made by FCC staff)

Ø When the appointment is made, offer the contact person the use of a small refrigerator for the duplicate food. This is preferred over storing the food in the household refrigerator. Mention that the urine samples will need to be refrigerated. On the Visit Form for the field staff, note whether a refrigerator is needed.

#### 6.1.2 Identification Labels

The Interviewer will:

Ø Make sure that the ID labels have the correct digits for the household, respondent, geographic stratum, and Cycle.

Ø Attach ID labels to the duplicate diet logsheet (3 parts) and chain-of-custody form. The sample type (digits 7 and 8) of the ID numbers should be 51 and 52, for duplicate diet (solid foods and beverages).

Ø Attach ID labels to the food checklist and its chain-of-custody form. The ID number should have sample type 07.

#### 6.1.3 Equipment for Day 1

Ø Select enough jars and bags to allow storage of four days' food and beverage consumption, as listed in section 5.1.1.

Ø Attach identical ID labels: sample type 51 to the bags, sample type 52 to the jars, and both to the refrigerator. If the respondent will be using a study refrigerator, pack the jars, and bags in the refrigerator, and the cold packs in its freezer. If the respondent will be using the household refrigerator, pack the jars, bags, and cold packs in a carton.

Ø Pack the equipment, including paperwork (section 5.2).

#### 6.1.4 Equipment for Day 8

Ø If the respondent is not using a project refrigerator, clean the cooler (with water and detergent) and air dry. Affix ID labels, sample types 51 and 52, to it.

#### 6.2 Sampling Location

The sampling location is the respondent's home. The refrigerator will be placed in a location convenient for the respondent, probably the kitchen or pantry.

#### 6.3 Sample Collection Procedure

##### 6.3.1 During Visit 1 (Day 1), the Interviewer will:

Ø Write the date and your name and signature on the logsheet.

Ø If the respondent will be using a study refrigerator, ask what would be a convenient location and put it there. Plug it in and make sure it is running. Show the target individual the bags and jars. Ask him or her to leave the cold packs in the freezer to keep the food cold after it and the refrigerator are collected on Day 8.

Ø If the respondent will not be using a study refrigerator, give the target individual the bags and jars. Ask him or her to leave the cold packs in the freezer of the household refrigerator to keep the food cold after it is collected on Day 8.

Ø Instruct the target individual that s/he is to keep a food checklist and collect duplicate food for all meals eaten for four days. This will include all food and beverages, whether eaten at home or elsewhere.

Ø The preferred days are Days 3-6 (e.g., Wednesday through Saturday if Day 1 is Monday). If the individual will be unable to collect duplicate food for a particular day or meal (e.g., if s/he is a dinner guest at someone else's home), s/he may substitute another day or meal. However, four full days' meals must be collected.

Ø Give the target individual a sheet of instructions for the duplicate diet collection, including a telephone number to be called if questions arise. Go through the instructions, making sure that s/he understands them.

Ø Instruct the target individual to collect a duplicate of each meal and snack s/he consumes during the four-day period. Include vitamin and mineral tablets and other nutritional supplements, and over-the-counter medicines (aspirin, antacids, etc.) but not prescription medicines.

- Ø Give the target individual the food checklist. Discuss the use of the checklist. Ask the individual for examples of foods s/he is likely to eat. If any are not on the checklist, discuss how to mark them.
- Ø Explain that the target individual should keep the food checklist for the same meals for which s/he saves duplicate food. S/he should write on the checklist the days or meals included.
- Ø Tell the target individual that on Day 5 or 6, someone will call to confirm the visit on Day 8, and will ask how many beverage containers and solid food bags s/he has used. At that time s/he can ask any questions s/he may have. If s/he has questions at another time, s/he may call the number on the instruction sheet.

#### 6.3.2 Interim Telephone Call to Respondent by FCC Staff

- Ø On Day 5 or 6, call the target individual to confirm the visit on Day 8. Remind him or her to take a urine sample on the morning of Day 8.
- Ø Ask whether s/he has any questions or problems, and how many solid food bags and beverage containers s/he has used.
- Ø Ask, in a nonjudgmental way, if duplicates of all foods and beverages have been saved. If not, ask which ones were not saved and why not. Record the answers on the logsheet, along with your name and signature and the date and time.

#### 6.3.3 On Day 8, before leaving the residence, the Field Interviewer will:

- Ø On the logsheet, write your name and signature and the date. Ask the target individual, in a nonjudgmental way, if duplicates of all foods and beverages were saved. If not, ask which ones were not saved and why not. Record the answers on the logsheet.
- Ø On the logsheet, note the number of food bags and beverage jars used in the four-day period.
- Ø If the respondent used a study refrigerator, make sure that all the collected food and beverages and the cold packs are in it.
- Ø If the respondent did not use a study refrigerator, pack all the collected food and beverages and the cold packs into the cooler.
- Ø Latch the cooler or strap the refrigerator to the handcart. Return the cooler or refrigerator to the FCC.

#### 6.4 Labeling of Samples

A unique ID number will be assigned for each sample (see HSPH SOP G03 "Identification Numbers for Samples and Forms"). Printed labels will show the ID number in bar-code and human-readable format. The interviewer will affix identical labels to each sample container, the refrigerator or cooler, the logsheet, and the chain-of-custody form.

The food checklist will be have affixed to it a label with a unique ID number, with sample type digits different from the duplicate food labels but the digits for household, individual, stratum, and Cycle the same.

#### 6.5 Preservation and Storage

Upon return to the Field Coordination Center, the Field Interviewer will:

- Ø Make sure that the ID labels on the samples in the refrigerator or cooler are identical.
- Ø Obtain a clean aggregate sample container. Peel the ID label from one of the sample containers and affix it to the aggregate sample container.

Remove this bullet

- Ø Weigh the aggregate sample container with its lid. Record the mass on the logsheet.
- Ø Open each solid sample container and inspect the food. If there are any inedible materials such as bones or orange peels, remove them. Scrape the food into the aggregate sample container. Add the snack food and beverages to the aggregate sample container. Put the lid on.
- Ø Weigh the full aggregate sample container with its lid. Record the mass on the logsheet.
- Ø Transfer custody of the aggregate sample to the Field Coordinator. Both Interviewer and Field Coordinator will sign the chain-of-custody form to record the transfer.

#### 6.6 Handling and Shipping

The aggregate sample will be stored in a refrigerator at 4°C while awaiting shipping to the FDA analytical chemistry laboratory.

Shipping to the FDA analytical chemistry laboratory will be on a timely basis with details to be worked out with the cooperating laboratory. Shipping should occur at least twice per week as storage of these bulky items is problematic.

Shipping will be via Federal Express or similar carrier. Samples will be shipped in protective containers with sufficient ice packs to maintain a temperature of 4°C throughout shipment.

## 6.7 Laboratory Analysis

The FDA laboratory will do extraction and analyze for pesticides and metals. If sufficient extract is available, FDA will send extract to HERL for PAH analysis. This is provided for in the EPA/FDA IAG.

A copy of the Standard Operating Procedures of the cooperating laboratory will be maintained on file by the Principal Investigator.

## 6.8 Data Workup

Field and laboratory data will be returned to Harvard in both magnetic and hardcopy format. Data will be coded and checked, computer entry verified, and discrepancies resolved. Analytical results will then be merged with questionnaires and other data, using the ID number as the merge parameter.

Results from this investigation will also be compared with results from the Mini-Market Basket survey. Mean pollutant concentrations will be determined for this study and a direct comparison made with concentrations from the Mini-Market Basket survey.

## 6.9 Sample Tracking

The ID number will allow tracking of each sample. A data base management system will ensure knowledge of the status and location of any sample at any time including retrospectively.

The chain-of-custody form will accompany the sample wherever it goes. Anyone who receives, transfers, or ships the sample will sign and date it, and keep a photocopy. It must clearly contain all necessary information so that the custody of the sample can be determined at any time. Airbills, bills of lading, etc., are acceptable substitutes when a commercial or government carrier is used; copies of such bills will be attached to the chain-of-custody form. Copies will be maintained at the Field Coordination Center. Copies of all records will be forwarded on a weekly basis to the Principal Investigator.

A copy of the chain-of-custody form will be sent to:  
NERL-CI

c/o Lisa Jo Melnyk  
26 W. Martin Luther King Drive  
Cincinnati, OH 45268

Phone: 513-569-7497  
Fax: 513-569-7115  
Email: [melnyk.lisa@epamail.epa.gov](mailto:melnyk.lisa@epamail.epa.gov)

## 7 Quality Assurance Procedures

### 7.1 Use of Laboratory and Field Blanks

No field or laboratory blanks will be used in this investigation.

### 7.2 Duplicate Sampling



Owing to burden on the respondent, no duplicate sampling will be done.

### 7.3 Quality Control

The Research Triangle Institute reports a sensitivity limit of less than 0.4 ng of lead per gram of food (Thomas, et al. RTI/5417/02/03 IG. Table 6-25). Other metals may be expected to be measurable at comparable levels.

FDA SOP 100 reports that recovery of laboratory spike sample and reference material is 70-130% for metals and pesticides.

### 7.4 Tolerance Limits, Detection Limits, and Sensitivity Limits

Category	Pollutant	Expected Concentration	Expected Limit of Detection Given Sampling Protocol
Metals	Arsenic	3.96 ng/g	3 ng/g
	Cadmium	20.1 ng/g	2 ng/g
	Chromium	1-2 ng/g	20 ng/g
	Lead	23.7 ng/g	10 ng/g
Pesticides	Chlordane	0.023 ng/g	1.5 ng/g (cis)
	Chlorpyrifos	0.111 ng/g	3 ng/g
	Chlorpyrifos-methyl	0.295 ng/g	1-5 ng/g
	4,4'-DDD		1-5 ng/g
	4,4'-DDE		2 ng/g
	4,4'-DDT	0.302 ng/g (total DDT)	3 ng/g
	Dieldrin	0.076 ng/g	1.5 ng/g
	Heptachlor	0.020 ng/g	1.5 ng/g
	Malathion	1.781 ng/g	3 ng/g
PAHs	Anthracene		
	Benzo(a)-pyrene		
	Chrysene		
	Phenanthrene		

## 8 References

Harvard University/Johns Hopkins University Standard Operating Procedures:

- G03 Identification Numbers for Samples and Forms
- G04 Chain-of-Custody and Sample Tracking
- G05 Storage and Shipping of Samples
- L06 Extraction of Metals from Sampling Media
- L07 Analysis of Metals by GF-AAS
- L08 Analysis of Metals by ICP-MS
- L10 Extraction of Neutral Pesticides and PAHs from Sampling Media
- L14 Determination of Pesticides, Acid Herbicides, and PAHs by GC/MS

Thomas, et al. *Draft Final Report. Measuring Personal Dietary Exposures: Phase I -- Pre-Pilot Testing of Duplicate Diet Guidelines.* Research Triangle Institute. RTI/5417/02-02 FR. September 1993.

Thomas, et al. Draft. *Guidelines for Measuring Personal Dietary Exposure to Environmental Contaminants.* Revision 1.0. Research Triangle Institute. RTI/5417/02-03 IG. September 1993.

U.S. Food and Drug Administration Standard Operating Procedures, May-July 1995:

- 100 Quality Control for Analysis of NHEXAS Food or Beverage Composites for Trace Elements and Pesticides
- 101 Determination of Pb, Cd, Cr, and Ni in NHEXAS Food or Beverage Composites by Graphite Furnace Atomic Absorption Spectrometry
- 102 Determination of Ba, Cu, Mn, V, and Zn in NHEXAS Food or Beverage Composites by Inductively Coupled Plasma Atomic Emission Spectrometry
- 103 Determination of As and Se in NHEXAS Food or Beverage Composites by Hydride Generation Atomic Absorption Spectrometry
- 104 Storage and Custody of NHEXAS Food or Beverage Composites
- 201 Sample Preparation in NHEXAS Food or Beverage Composites by Homogenization
- 202 Determination of Selected Organohalogen and Organophosphorus Residues in NHEXAS Solid Food Composites
- 203 Determination of Selected Organohalogen and Organophosphorus Residues in NHEXAS Beverage Composites

## **NHexas Duplicate Diet Study -- Instructions for Participants**

The purpose of this study is to collect duplicates of all the food you eat for four days. This food will be analyzed for lead and other metals, pesticides, and PAHs (polycyclic aromatic hydrocarbons), which are found in smoke. You will also keep a food checklist for the same four days. Both the duplicates and the checklist will include all food and beverages, whether eaten at home or elsewhere.

### **Duplicate Diet Instructions**

Choose four days on which you will collect duplicate food. It may be most convenient to choose days 3-6 (Wednesday through Saturday if Day 1 is Monday). If you will be unable to collect duplicate food for a particular day or meal (for example, if you are a dinner guest at someone else's home), you may substitute another day or meal. However, four full days' meals must be collected.

Collect a duplicate of each meal and snack you consume during the four-day period. The meal-preparer will need to cook enough food to allow for the extra plate to be served. If you eat at a restaurant, order two identical meals. Include vitamin and mineral tablets, other nutritional supplements, and over-the-counter medicines (aspirin, antacids, etc.) but not prescription medicines.

Serve the duplicate food into dishes and cups like those for the consumed meal (or as alike as possible), in order to compare quantities. Leave the food in the dishes and cups until you finish the meal. If you take another helping, add the same amount of the same food to the duplicate dish. If you do not finish a food, remove the same amount from the duplicate dish before you store the food.

Remove inedible material (such as bones, eggshells, and orange peels) from the duplicate food. Remove any material that you do not eat; for example, if you do not eat the skin of a baked potato, remove the skin from the duplicate food.

Place solid foods in a plastic bag. Seal the bag and place it in the refrigerator. There are five bags for each day. You may find it most convenient to use a different bag for each meal or snack. It is not necessary to keep meals separate, however. If you prefer to put more than one meal into a bag, or if you have a large meal and put some of the food into another bag, this is fine.

Pour beverages, liquid foods (such as soups), and foods that may melt (ice cream, etc.) into the beverage jars. Tighten the lids securely and store the jars in the refrigerator. Place drinking water in a beverage jar as well even though a separate sample of drinking water will be taken. To save refrigerator space, empty jars and jars containing only

water may be stored outside the refrigerator.