Tracer Applications in the California Regional $PM_{2.5}$/PM_{10} Air Quality Study

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Table of Contents

1. Introduction

1.1 Report Objective

The objective of this report is to examine potential tracer applications for the forthcoming California Regional $PM_{10}/PM_{2.5}$ Air-Quality Study (CRPAQS), with specific attention to the following elements:

- potential uses of tracer studies in the context of the comprehensive CRPAQS; and

- expected scientific returns of such studies, as well as their cost-effectiveness and probabilities of success.

This report fulfills this objective by sequentially examining several possible goals of candidate tracer application within the CRPAQS context, identifying salient operational and scientific aspects of each, screening these applications according to their relevance to CRPAQS goals and technical feasibility, and selecting a short list from these candidates that is recommended for further consideration. Final sections of this report examine applications from this shorter list in greater detail, with the intent of providing CRPAQS planners with launch points, should any of these candidates be chosen for deployment.

1.2 Bounding Conditions: Relation to the CRPAQS

Scientific planning for non-tracer aspects of the CRPAQS is currently well underway, with formal documentation of science objectives, and definitive plans for the associated field and modeling efforts (Watson, et al. 1998). Because of this advanced planning state of the core program and also because of associated timing considerations, this tracer-application report is bounded by three important constraints, which are listed as follows:

- The tracer applications suggested here must serve, and promote attainment of, the key objectives currently set forth for the overall CRPAQS.

- These applications must provide a reasonable complement to the planned ensemble of CRPAQS field-observation facilities, using these facilities and the resulting data sets to the maximum practical extent that is commensurate with CRPAQS objectives. This implies strongly that the timing of any tracer measurements should be concurrent with those of the major field studies.

- The short time-frame and limited nature of this tracer-application report precludes in-depth design. Consequently, this report will provide only semi-quantitative aspects of associated tracer deployment, measurement, and interpretive features. Complete, quantitative specification of these features should follow if and when the CRPAQS community chooses specific tracer application(s).

1.3 Tracer Studies: Some General Considerations

A number of atmospheric tracer studies conducted during past years have produced highly valuable and insightful results, while others have fallen well short of their intended goals. When failures do occur, they generally stem from one or more of four principal causes, namely:

1. invalid, or non-robust conceptual design;
2. inadequate sampler coverage (i.e., sampler network spacing, areal extent, time-resolution, and/or duration) for quantitative documentation of tracer fields;
3. inadequate quality control of tracer-release, sampling, and/or analytical procedures; and
4. circumstantial elements, such as inopportune weather conditions, equipment malfunctions, and related considerations.

Numerous illustrative examples can be given for each of these. Conceptual design flaws can occur when tests (such, for example, as vertical pollution-transport studies) are designed on the presumption of low-dimensional behavior, but when in reality higher-dimensional components occur. Other design deficiencies occur frequently when tracers are released with presumably co-located pollution sources to examine the fate of that pollution, only to discover that differences in the time-histories of release or that extraneous pollutant sources have clouded interpretation of the results. Such circumstances often have occurred in hindsight, leading to the observation that tracer studies often "look far better at first glimpse than they do after extended scrutiny." Because of these and other potential pitfalls, careful initial attention to conceptual design, as well as the more operational aspects, is warranted.

Even with a valid conceptual design, practical features have often arisen to cloud the interpretation and value of tracer results. As indicated above, sparse sampler coverage — which usually results from the expense of deploying widespread sampling arrays — often prevents "closure" of the fields under observation and thus leads to results of diminished interpretive value. Quality control, which is difficult to maintain at the low concentration levels typical of tracer application, is another issue. Although several tracer studies have included excellent levels of quality control there are notable exceptions, where limitations in measurement reliability have precluded almost all useful application of the resulting data.

The concept of closure is useful in examining this and other components of the above list. In the strongest possible sense, "closure" describes the ability of a study's design to account quantitatively for all elements of the tracer's material balance, using detailed flux measurements taken at downwind points. This is invariably a difficult feat in a three-dimensional, time-variant world; consequently, many studies have been designed to avoid the necessity for closure. Still others accept a weaker sense of the term, which allows quantitative capture of only selected components of the material balance (such as the deposition field of a depositing tracer). In an even weaker sense, several tracer studies have been content to measure a "footprint", such as a surface concentration field, that relates to the material balance only indirectly, often using dispersion models to infer other features. The important element in both "strong" and "weak" closure cases is the observational system's ability to quantitatively capture the total field of interest: Even weak closure is not attained if some of the field escapes the observation system and thus precludes quantitative measure.

Within this context, it is useful to categorize tracer studies into the four broad classes, which attempt to deal with closure in different ways, as indicated in Table 1.1. These categories will be referenced periodically in the candidate applications discussed in the following sections.

1.4 Report Organization

Because of the above-noted need to mesh the content of this report with the established objectives and field-study design of the CRPAQS, the following section will present a summary of key features of the CRPAQS core program for downstream reference by later sections. Section 3, which follows immediately thereafter, provides an overview of a number of potentially applicable tracer types, giving sufficient detail to examine field-study feasibility and to point the reader to more detailed literature in the field. This is followed in turn by Section 4, which outlines a list of possible objectives for tracer deployment, and links these with the tracer types and techniques that are most applicable for their specific purposes. Section 5, which constitutes the most important element of this report, distills the objective list from Section 4 into six candidate objectives. These are discussed sequentially, giving relatively in-depth examinations of their relation to CRPAQS objectives, potential scientific return, probability of success, and cost-effectiveness. Section 6 concludes the report, listing several general recommendations for further action by the CRPAQS, which are intended to guide future strategic planning associated with tracer applications.

It should be noted that although a large number of past tracer studies are cited in the reference material, provision of an explicit, comprehensive review of past tracer studies is not an objective of this report, and consequently such a review does not appear here. Salient information gleaned such studies is, rather, incorporated in a more implicit fashion in the prospective analysis of the selected objectives, which appears in Sections 4 and 5.

Table 1.1: Broad Categories of Tracer Studies

Class of Study	Closure Requirements	Examples
1. Quantitative material balances and "footprint" measurements. *"Closure"*	Attempt to attain closure in the strong or (usually) the weaker senses.	*Strong sense:* Hanford ^{85}Kr diffusion studies (Nickola 1977); *Ranging from weak sense to incomplete closure, depending on coverage of individual test:* Long-Range Studies: CAPTEX (Ferber, et al. 1986), ANATEX Draxler, et al. 1991), Mohave Pitchford, et al. (1997), ETEX (1997), SJVAQS (Tracer Technologies, 1991; Lehrman, et al. 1994)). Scavenging Studies: (Gatz, 1977). Complex Terrain Studies: ASCOT (Clements, et al. 1989)
2. Partial measurement of material-balance components. *"Incomplete Closure"*	Accept partial field measurements. Typically produces qualitative results only, unless supported by modeling analysis.	
3. Hypothesis testing ("if hypothesis A is true, then we should expect to see tracer at point x and time t"). *"Hypothesis Testing"*	Ignore closure considerations completely; typically produces qualitative results only.	Vertical Transport Studies: Ching and Alkezweeny (1986)
4. Ratioing measure-ments. *"Ratioing"*	Avoid necessity for closure by ratioing tracer with one or more pollutant constituents.	Plume-Decay Studies: (Doran and Horst, 1985, Kallend, 1983, Easter, et al. 1980). Source-Apportionment Studies (Green and Tombach, 1997)

2. CRPAQS Overview

As noted above, the CRPAQS objectives and field-study design are currently at a well-defined stage. The following subsections summarize pertinent aspects of this program to an extent sufficient to launch the later discussion of possible tracer applications. This is an intentionally abbreviated account, and additional, more detailed information will be employed periodically throughout this report as required. This more extensive information is available in the plan by Watson, et al. (1998), which can be downloaded from the ARB Web site at <http://sparc2.baaqmd/centralca/>.

2.1 Field-Program Timing and Locations

Timing

The CRPAQS field effort is divided into a "Long-Term Campaign" extending between 1 December 1999 and 15 February 2001, a "Fall Campaign" occurring between 15 September and 15 November 2000, and a "Winter Campaign" scheduled for 1 December 2000 through 15 February 2001. Choices of fall and winter for intensive campaigns are based primarily on the observation of high aerosol concentrations during winter and (to a somewhat lesser extent) fall, combined with distinctly different meteorological regimes in force during these two periods (see discussion of scenarios in Section 2.2).

Locations

Figure 2.1 shows approximate locations of CRPAQS study-domain borders, to facilitate later discussion of potential tracer applications. More detailed maps are available in the Watson, et al. (1998) reference.

Modeling Domain:
Lines of roughly constant latitude and longitude, with the following approximate boundary locations bound the CRPAQS modeling domain:

Northern	Chico, California
Southern	Corona, California
Eastern	Barstow, California
Western	200 km W. of San Francisco

Large Domain:
Sampling systems within the "Large Domain" are within the Modeling Domain and are contained within the State of California.

Annual and Winter Study Domain:
This sub-domain of the Large Domain encloses the San Francisco Bay area as well as the San Joaquin Valley and the southern portion of the Sacramento Valley.

Fall Study Domain:
This small sub-domain of the Large Domain is situated in the San Joaquin Valley, including Fresno and a portion of the Western Sierra slopes.

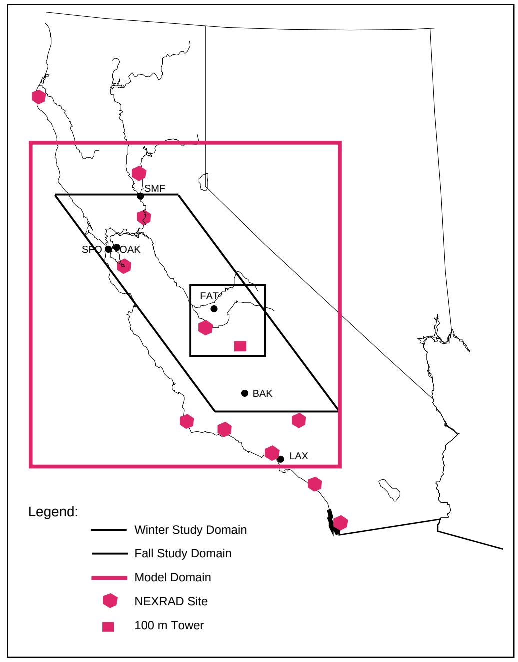

Figure 2.1: Approximate Locations of CRPAQS Study Domains and NEXRAD Radar Sites

2.2 CRPAQS Objectives

CRPAQS Programmatic Goal

The CRPAQS Programmatic Goal is *to provide additional and more comprehensive information than is currently available to explain the nature and causes of particulate concentrations and visibility impairment in and around Central California.* This new information is deemed necessary to better understand the outcomes of currently planned emission-control strategies, and to promote efficient, cost-effective selection of future strategies.

Specific Field-Study Objectives

Specific Objectives of the CRPAQS field program appear in the following list.[1] Included on this list are check marks indicating, on a scale of zero to three, an initial qualitative assessment of the potential usefulness of tracer measurements for each objective. More quantitative screenings of this type appear in later sections of this report.

1. ✔✔ Obtain a documented data set, with appropriate data qualification statements, that is suitable for characterizing the nature and causes of particulate concentrations and visibility impairment in and around Central California by supporting modeling and data-analysis activities.

[1] A more recent listing of objectives became available during the final preparative stages of this report. These "alternative objectives" are listed for reference in the text box on page 2-4.

2. ✔ Evaluate the extent to which long-term PM monitoring networks represent the levels to which large populations are exposed under a variety of emission and meteorological conditions.

3. Document the current spatial distribution, temporal variation, and intensity of PM concentrations and visibility impairment within Central California.

4. ✔✔✔ Measure and characterize the structure and evolution of the boundary layer and the nature of regional circulation patterns that determine the transport and diffusion of atmospheric contaminants in Central California.

5. ✔ Further characterize the source zones of influence and quantify source contributions to community exposure for PM chemical components, including particles that are directly emitted and those that form from directly emitted gases.

6. ✔ Quantify source contributions to secondary aerosol, identify the limiting precursors, and assess the extent to which reductions in nitrogen oxides, ammonia, sulfur oxides, and volatile organic compounds would be effective in reducing $PM_{2.5}$ concentrations.

7. ✔✔ Refine conceptual models that explain the causes of elevated PM concentrations, and interactions between emissions, meteorology, and ambient PM concentrations.

8. ✔✔ Evaluate and improve the performance of emission, meteorological, and air-quality simulations. Apply simulation methods to estimate PM concentrations at receptor sites and to test potential emission-reduction strategies.

Conceptual Models

In addition to stating the programmatic objectives noted above, the CRPQAQS panning documentation lists four broad "conceptual models" describing avenues leading to locally high concentrations of both fine and coarse aerosol particles, and sets forth multiple research questions designed to test these models. Although the research questions are too extensive to reproduce here, it is useful to cite the conceptual models for future reference in evaluating potential tracer applications. These are:

Alternate Statement of Objectives

1. CHARACTERIZATION OF PM

- Characterize ambient $PM_{2.5}/PM_{10}$ throughout the study: concentrations, chemical composition, and size distributions, including seasonal, temporal, and spatial variability.
- Characterize meteorological conditions associated with high PM concentrations.
- Characterize visibility, including seasonal, temporal, and spatial variability.
- Develop guidelines for assessing the extent to which adverse PM episodes are meteorologically driven vs. emissions-driven.

2. ATMOSPHERIC PROCESSES CONTRIBUTING TO PM FORMATION

- Develop a better understanding of key chemical and physical processes that contribute to elevated PM concentrations.
- Determine which precursors (VOC, NO_x, NH_3, HNO_3, SO_2) limit the formation of secondary aerosols, as a function of location and time of day.

3. EMISSIONS ESTIMATION AND VERIFICATION

- Develop reliable estimates of PM, VOC, NO_x, and NH_3, emissions. For PM, determine chemical composition and size characteristics, as well as emissions rates. For VOCs, determine chemical composition.
- Explain discrepancies between emission inventories and ambient measurements with respect to the relative amounts of PM derived from geological and combustion sources.
- Understand the role and contributions of biogenic emissions to secondary organic pollutant formation in central California.

4. TRANSPORT AND RELATED IMPACTS

- Determine the extent of transport of precursors and secondary pollutants between the San Joaquin Valley (SJV) and major California air basins - the Bay Area, the North Central and Central Coast areas, the Sacramento area, the southeast desert area, and the Sierra Nevada – and the contributions of these transported pollutants, by area of origin, to ambient PM concentrations in the receptor areas during each season of interest.
- Estimate the contributions of emissions from one portion of the southern Central Valley to ambient PM concentrations in other portions, where the Valley is divided into the Sacramento area, the North SJV (Modesto, Stockton), central SJV (Fresno), and south SJV (Bakersfield).
- Estimate the contribution of pollutants transported from the SJV to visibility impairment in the southeast desert and in Class 1 areas, notably national parks and forests. Estimate the impacts on visibility impairment in these areas resulting from possible emissions reductions in the SJV.
- Develop procedures for determining, for non-attainment areas, the upwind extent of the source region that should be subject to emissions reductions (i.e., the "zone of influence" issue) and its variation with chemical constituent and meteorological regime.

5. MODEL ADAPTATION AND EVALUATION

- Assess the degree of reliability of models for estimating ambient pollutant concentrations.
- Establish modeling capabilities for use in reliably estimating future air quality for hypothetical scenarios.

6. EMISSIONS REDUCTION REQUIREMENTS AND IMPACTS

- For monitoring sites located in central California having exceedances of the 24-hour and/or annual standards, determine which categories of sources contribute significantly to ambient concentrations, the relative proportion of their contributions, and the anticipated benefits of emissions reductions.
- Estimate the impact on ambient ozone concentrations of emissions reductions that are contemplated for reducing PM concentrations, and vice versa.

7. REGULATION-RELATED CONCERNS

- Determine the effects of meteorological variability on the likelihood of exceeding the standards. Assess the likelihood of "flip-flopping" into and out of attainment of the standards.

- Determine the extent to which high 24-hour average values contribute to exceedance of the *annual* average standard, regardless of whether the 24-hour average standard is exceeded. Assess the relative impacts of types of episodes on the annual average.

Primary avenues for high PM2.5 occurrences:

CM-1. Winter: secondary aerosol formation in fog, superposed on primary aerosol

CM-2. Fall: secondary aerosol formation by dry processes, superposed on primary aerosol

Primary avenues for large particle occurrences, which contribute to high PM10 concentrations:

CM-3. Low winds, with nearby man-made dust sources

CM-4. High-wind dust suspension

Although these conceptual models require little explanation, it should be noted that CM-1 and CM-2 incorporate the notion of high-pollution incidents being associated with synoptic high-pressure systems that impose strong inversion layers, effectively constraining pollution within the valley domains. Removal of particulate pollution is limited primarily to deposition and revolatilization processes under these circumstances. Major cleansing occurs periodically, when energetic synoptic systems move in to displace the high-pressure regions and, essentially, flush the pollutant from the valley environs. Quite obviously, any violations of these conceptual assumptions, particularly those involving confinement by the inversion layer and surrounding terrain, will have strong bearing on in-valley residence times and associated particle concentrations, and thus are of high interest in the CRPAQS effort.

2.3 Field Measurement Summary

Measurement equipment for the Long-Term, Fall, and Winter campaigns is summarized in the three following text boxes. Again the reader should note that this summary is intended solely to support the discussion and thus is intentionally brief. The Watson, et al. manuscript can be consulted for further details.

2.4 Model-Application Summary

Noting that "no single model will be adequate to address all aspects of PM" the CRPAQS plan lists a number of model categories for deployment during the time-frame of the study. These include, but are not limited to: • conceptual models, • emission models, • meteorological models, • chemical-transport models, and • receptor models.

Long-Term Campaign

Aerosol Measurements

- ARB Backbone $PM_{2.5}$ Network:
  - 45 24-hour samplers dispersed throughout Large Domain - 12 of these will operate continuously, 39 will operate every third day. All will sample aerosol mass; a subset of 27 will sample for element, ion, and carbon analysis every 12th day.

- ARB PM_{10} Network:
- 109 24-hour samplers dispersed throughout the Large Domain.

- CRPAQS Anchor Network:

Three sites (near Bakersfield, Fresno, and Angiola) sampling:
PM-10 mass
PM-2.5:
mass
light scattering
light absorption
carbon

- CRPAQS Satellite Network:

Satellite sites located at key positions associated with transport, gradients, and emissions, acquiring:
PM-2.5 samples amenable for chemical analysis
Nephelometer data

- Instrumented 100 m tower, located at the non-urban Angiola site (see Figure 2.1):
Continuous, automatic particle-size instrumentation at 5 levels.

General Air-Quality Measurements

- Several sites distributed throughout the Large Domain:
134 ozone
78 NO_x
33 SO_2
16 light scattering
47 light absorption

Meteorological Measurements

- Surface Meteorology:

Combination of eight networks throughout Large Domain, including
157 wind speed and direction sites
122 temperature sites
60 humidity sites
26 solar radiation sites
7 barometric pressure sites

- Upper-air meteorology:
Array of profiler, RASS and Doppler sodar sounders (1-hour observations)
Rawinsondes at Vandenburg, Oakland, Point Mugu, and Edwards (Twice-daily launches)

- Instrumented 100 m tower, located at the non-urban Angiola site (see Figure 2.1):
5 levels of wind speed, temperature, and direction

Fall Campaign

Aerosol Measurements

- Network enhancement

The ARB $PM_{2.5}$, ARB PM_{10}, and CRPAQS networks described above will be enhanced by increasing sampling frequencies and adding monitors at some locations.
The CRPAQS Anchor Network will include 24-hour PM_{10} measurements on substrates amenable to the identification of different dust types, particle morphology, ions, carbon, and elements.

General Air-Quality Measurements

- Ammonia influences on particle formation

Continuous NO_3, SO_4, HNO_3, and NH_3 measurements will be conducted around a large rural NH_3 source near Fresno to examine HNO_3/NO_3 gas/aerosol relationships and the associated influences of NH_3.

Winter Campaign

Aerosol Measurements

- CRPAQS Network enhancement

The CRPAQS Anchor Network will be enhanced by installing similar sampling equipment at the Bethel Island site (for evaluation of interbasin transport) and a new Sierra foothills site (near the top of the wintertime valley-wide surface layer). Continuous monitoring for NO_3, SO_4, HNO_3, and NH_3 will be conducted at all five sites.
The Satellite Network will operate continuous, 24-hour sampling for chemical speciation, and will add backup filters for HNO_3, and NH_3. Five additional satellite sites will be operated in source-dominated areas.

- ARB Backbone $PM_{2.5}$ Network enhancement

All sites on the ARB PM_{25}. network will be operated on a continuous basis during 20 days of the Winter Campaign.

The CRPAQS plans to conduct grid-based meteorological and chemical-transport modeling on a daily basis for a full year during the Long-Term campaign, with a subset of about 60 days to be examined in greater detail. These grid-based modeling activities will be enhanced further during the Fall and Winter campaigns, with features such as enhanced spatial resolution and more detailed chemical and microphysical mechanisms.

Receptor models will be applied during the Long-Term, Fall, and Winter campaigns, as appropriate to the suite of measurements obtained during these periods. More detailed discussion of modeling plans and approaches is available in the Watson, et al. manuscript.

3. Overview of Candidate Tracer Types

Although inert gaseous tracers such as SF_6 and perhalogenated organics represent a major class of tracers anticipated for application in the CRPAQS, a variety of other tracer types, such as fluorescent compounds, rare-earth elements, radar reflectors, and natural "tracers of opportunity" are of interest as well, either as single tracers or as companion components to investigations where perhalocarbons are employed. Many of these additional tracers are aerosols, thus providing the possible advantage of mimicking, at least approximately, the depositional behavior of their pollution counterparts. This section summarizes briefly the components of this potential "tracer arsenal," to provide an information backdrop for the subsequent discussion.

3.1 Perfluorinated Organics

Perfluorinated tracers have been used extensively in past California field studies, and the CPRAQS team is familiar with their use, their potential, and the pitfalls associated with their application. Accordingly, this discussion will be centered mainly on recent advancements in this field, which promise to enhance the economy, versatility, and interpretive applicability of these materials.[2] These advances have centered mainly on improvements of specificity and sensitivity in the detection technique, and the development/evaluation of new tracer compounds. These are discussed sequentially in the following paragraphs.

Sensitivity/Specificity

Specialized column-chromatography techniques developed over the past few years have lead to substantial sharpening of eluent peaks, resulting in marked improvements in both sensitivity and peak resolution. In particular, the Carbon Layer Open Tubular (CLOT) technology provides sufficient resolution to separate ortho, meta, and para cis/trans isomers, as well as different molecular-weight compounds, and leads to sensitivities in the range of a few tenths of a femtogram. Such sensitivities allow precise determination of the more conventional tracers at their global background concentrations. Since these global values are known and quite uniform spatially, this provides an internal QA standard for such measurements. Quite obviously, these increased sensitivities decrease the amount of tracer-release necessary for detection at remote locations downwind, thus enhancing the economy of tracer application.

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[2] The author expresses his appreciation to Russel Dietz of Brookhaven National Laboratory, who has supplied much of the information contained in this subsection.

Fast-response, mobile perhalocarbon monitors currently are experiencing a similar advancement. At present Brookhaven Laboratory is completing a CLOT-based mobile monitor which is suitable for aircraft deployment, and should have peak-separation features similar to those noted above and a response time on the order of 30 seconds.

To provide some indicators for scoping purposes, one can estimate roughly that releases of 0.1 kg/hr tracer should be detectable at 1000 km distances at the plume's ground-level centerline, given the above-noted sensitivities. Costs for the more conventional perfluorocarbon tracers, normally purchased from in England, are roughly $400.00/kg. An exception is perfluorodimethylcyclobutane (PDCB), which is manufactured by DuPont and costs about $50.00/kg. Deployment costs for automated bag or adsorption-trap samplers can be estimated at approximately $100.00 per sample, with analysis costs also falling in the $100.00 per sample range. Deployment costs for passive adsorption-trap samplers will be substantially less than for their automated counterparts.

New Perhalogenated Tracer Compounds

Currently a number of additional perfluoronated tracer compounds are being tested, which are candidates for addition to the existing arsenal. The main advantage of these additional species relates to their low global backgrounds, which permit unambiguous source detection at levels even lower than their more conventional counterparts. Table 3.1 lists some of the compounds currently being tested in a DOE-sponsored effort at Brookhaven National Laboratory, along with some of the more conventional species. Ambient concentrations for the new compounds under test are typically of the order of a few tenths of a part per quadrillion (ppq), compared to about 5 ppq for the more conventional "standard" tracers.

Table 3.1: Some Perfluorocarbon Tracers

"Standard" Perfluorocarbon Tracers
Perfluoromethylcyclohexane (PMCH)
Ortho- and meta-perfluorodimethylcyclohexane (o, mPDCH)
Perfluoromethylcyclopentane (PMCP)
Perfluorotrimethylcyclohexane (PTCH)

Potential Additional Perfluorocarbon Tracers
Perfluoroethylcyclohexane (PECH)
Normal perfluoropropylcyclohexane (nPPCH)
Iso perfluoropropylcyclohexane (iPPCH)
Ortho-, meta-, and para-perfluormethylethylcyclohexanes (o, m, pPMECH)
Perfluorodimethylcyclobutane (PDCB)
Perfluorotrimethylcyclohexane (PTCH)

3.2 Additional Nonreactive Gaseous Tracers

Several possibilities exist for observing additional nonreactive gaseous tracers, including both tracers that are intentionally released as well as "tracers of opportunity," which are emitted through human activity or by natural processes. The work by Miller, et al (1990) (see also Bastable, Schorran, and Rogers, 1990) provides an example of the latter type. These authors measured aerosol properties concurrently with observations of trichlorofluoromethane, carbon tetrachloride, methylchloroform, and perchloroethylene at an elevated Southern Nevada site, using a ratioing technique to infer contributions to visibility degradation from the Los Angeles basin.

Several additional nonreactive gases are available for intentional release, which can be used as conservative tracers Dietz (1986). Including species such as sulfur hexafluoride and deuterated methane, these compounds are generally less attractive than perfluorocarbons for the CRPAQS because of exotic preparation/analysis requirements, reduced detection sensitivity, or both. The reader is referred to the Dietz review for further details on this subject.

3.3 Fluorescent Particles

Fluorescent dye particles have been applied over the past few decades for a number of atmospheric studies, including measurements of plume diffusion (Draxler, 1984), dry deposition (Sehmel, 1984), precipitation scavenging (Englemann, 1965), and particle resuspension (Sehmel, 1984). Detection of the tracer, which may take place either *in situ* or on a collected sample, occurs by observing fluorescent radiation stimulated by electromagnetic or sub-atomic particle irradiation of the tracer.

Bulk Sampling Techniques

The preponderance of past studies employing fluorescent particles has involved bulk releases of inexpensive, polydisperse dye aerosols such as uranine, rhodamine, and zinc sulfide, with subsequent filter or environmental-medium (e.g., soil, rainwater, . . .) collection. Owing primarily to sensitivity limitations, distance scales of these studies have been limited to a few kilometers or less (meso-gamma scale). Quite obviously, deployment of dye aerosols in this mode cannot be considered a substitute for the longer-range gaseous tracers noted above.

Such applications still may be attractive, however, depending on the specific objectives of a particular CRPAQS field-study component (such as the measurement of deposition rates), which may be more short-range in nature. On the other hand, uncertainties associated with the polydispersity of the aerosol, and possible ambiguities in the relationship between the tracer and the aerosol that it is intended to represent, are potential drawbacks that must be considered in the design of any such application.

Single-Particle Techniques

More recent, *in situ* techniques provide the possibility of extending the applicability of fluorescent-particle tracers appreciably beyond the limited scope indicated above. One such technique, for example, consists of adapting a conventional single-particle optical counter to incorporate a monochromatic light source, which is tuned to a specific wavelength to activate the dye compound in question (Harrison and Lin, 1992). Such counters can provide unit counting efficiency for particles down to 0.1 microns diameter and, moreover, are able to size as well as count particles. This enables extremely sensitive observation of modest releases of inexpensive tracers at downwind distances comparable with those normally associated with perfluorocarbon tracers, at substantially improved time-resolution. Moreover, the sizing capability of the system suggests that one can use polydisperse tracer releases to advantage, relating deposition rates to relative concentrations of different-sized particles as they are observed at increasing downwind distances. One interesting adaptation of this method involves the generation of special, dye-containing "designer" aerosols that are created to have special properties: for example non-wettable aerosols consisting of dyes encapsulated in paraffin, and hygroscopic dye-containing NaCl particles.

There are several potential practical limitations associated with this technique in the CRPAQS context. First, and although the method has been demonstrated to be workable, a fleet of deployable counters does not exist at the present time. Unit price is somewhat expensive ($20K estimate), and construction time could be a major logistical factor. In addition, some evidence exists that coagulational adducts of the tracer particles with natural aerosols may enhance florescent response, thus giving skewed particle-sizing results. On the other hand, the substantial attractiveness of the features noted above, combined with the potential for deployment on mobile platforms, places this methodology rather high on the interest list for CRPAQS application.

Fluorescent-Particle Lidar[3]

Past studies have demonstrated that lidar units, with receivers tuned to the emission wavelength of fluorescent dye particles, can successfully trace experimental dye releases for several hundred kilometers (Johnson, 1985; Uthe,

[3] Conventional lidar is incorporated into the base CRPAQS field plan, and will not be addressed in this report. Any comments made here pertaining to fluorescent-particle lidar should not be construed as pertaining to other lidar applications.

1991; Uthe, et al. 1985). Such "FP-lidar" applications have two substantial advantages over perhalogenated, and most other, inert gaseous tracers: First, the fluorescent-particle tracer is relatively inexpensive; second, the technique provides three-dimensional remote sensing on a highly time-resolved basis. Possible disadvantages include the somewhat more cumbersome nature of the tracer-release methodology, and dry- and wet-deposition of the tracer, which will render it progressively nonconservative with increasing downwind distances. FP lidar is essentially un-usable during fogging conditions.

Development and application of tracer technology using FP-lidar appears to have slowed appreciably during the past decade, and thus is not at a readiness state comparable to perhalocarbon technology. Many of the existing elastic-scattering lidars can be adapted for FP application relatively easily, however, should this need arise.

3.4 Rare Earths

As their name implies, rare-earth elements such as osmium, tantalum, indium, and europium have low crustal abundances and therefore offer the possibility of unambiguous detection as tracers at low ambient concentrations. Such tracers are typically generated as aerosols, either by ignition of specially prepared flares or by combustion of sprays of organic liquids containing dissolved ligands of the selected element(s). Such dispersion techniques typically result in wide particle-size distributions, which are rather difficult to control and characterize in a reliable, quantitative manner. Chemical analysis of recovered samples is usually performed using neutron-activation, or other techniques involving rather specialized analytical procedures. Analytical costs can be estimated nominally at $100.00 per sample.

Although potentially useful for a variety of tracer applications, rare earths have been applied most frequently for measurement of in-cloud scavenging rates and processes, where the tracer is dispersed in the storm inflow or cloud regions, with subsequent recovery and measurement of tracer in collected precipitation samples. For air-motion tracing and/or deposition tracing these tracers appear to have little or no advantage over other methods, and thus are of marginal interest to the CRPAQS.

3.5 Natural Radionuclides

Although a number of natural radionuclides, including cosmogenic and crustal species, have been applied for as "tracers of opportunity" for various atmospheric-science applications (e.g., Kritz, Roulley, and Danielsen, 1990;

Langner, Rhode, and Olofsson, 1990; Cohen, et al. 1972; Lee and Larsen, 1997, Butterweck, et al. 1994; Guedalia, et al. 1974; Shaffer, 1973), radon and its daughter products appear to be of highest potential value in the CRPAQS context. There are several reasons for this. First, radon originates from the surface, providing advantages over species from cosmogenic sources such as ^{7}Be, which have no obvious or direct applicability to field studies such as this. Furthermore, as a decay product of crustal uranium, radon emissions are distributed more-or-less uniformly over land surfaces and, at least for long-term averages, are quite constant in time. Atmospheric concentrations of radon, ranging between 100 Ci/m^3 up through several thousand Ci/m^3, fall well within a range that is measurable using standard techniques (Intersociety Committee, 1972). Finally, radon decays in a chain sequence to form a sequence of products having a variety of lifetimes; i.e.,

$$^{222}Rn \xrightarrow{\alpha,\ 5.49\ Mev;\ 3.8235 days} {}^{218}Po \xrightarrow{\alpha,\ 6.00\ Mev;\ 3.05 min} {}^{214}Pb \xrightarrow{\beta,\ 0.22\ Mev;\ \gamma,\ 0.23\ Mev;\ 26.8 min}$$

$$^{214}Bi \xrightarrow{\beta,\ 0.63\ Mev, \gamma,\ 2.03\ Mev;\ 19.9\ min} {}^{214}Po \xrightarrow{\alpha,\ 7.69\ Mev; 0.0001\ sec} {}^{210}Pb$$

thus (under some rather idealized atmospheric conditions) radon and its progeny offer the possibility to clock, and thus measure the rates of, specific transport processes such as vertical exchange.

Radon and its progeny are measured by radioactive counting of either radon and/or its daughter products subsequent to collection in adsorbant traps, on filters, or as gas samples. The short half-lives of key radon progeny necessitate onsite analysis in most situations. Basic radon monitors that perform collection and counting in integral units suitable for field deployment can be obtained for between two and ten thousand dollars, depending on the vendor and the sophistication of the device and the specificity of the technique chosen. Obviously, more detailed analysis of specific daughter products will lead to more useful timing information concerning the transport processes being observed.

In addition to fixed samplers, current technology also allows airborne monitoring with temporal resolution in the range of ten minutes or less. Apparently the currently most advanced sampler of this type was constructed by the DOE Environmental Measurements Laboratory. This device utilizes an alpha-spectrometer to measure ^{218}Po and ^{214}Po, inferring radon concentrations based on the assumption that decay equilibrium exists. Applications of this technique for vertical-transport measurements are examined further in Sections 4.4 and 5.4.

3.6 Radar Reflectors

Radar reflectors, especially chaff, have the distinct advantage of providing remotely sensed estimates of air motions or even quantitative measures of three-dimensional wind fields, if two synchronized Doppler radars are employed. Vector plots of wind fields observed over the Santa Barbara coast during the SCCCAMP campaign, using the NOAA dual-Doppler facility (Wilczak, Dabbert and Kropfli, 1991) provide a vivid example of the later case in the California context. Quantitative and detailed three-dimensional wind-field information, in well-chosen atmospheric volumes and observation times, would be of obvious and high value for quantifying key aspects of atmospheric-transport behavior in the CRPAQS.

Although single radars cannot directly generate three-dimensional vector descriptions of wind fields, they still can be useful for simply following the transport and dispersion of well-defined and localized targets such as chaff, or tracers of opportunity such as dust, insects, and pollen. Moreover, if such radars have Doppler capability, they can at least provide measurements of radial velocity components and — under ideal conditions — mean winds.

Transportable research radars for both single and dual-Doppler application are available and can be located at field sites chosen for specific CRPAQS purposes (Kropfli and Kelly, 1996). In addition, the recently established array of WSR-88D "NEXRAD" weather radars (Crum, 1995; Crum, Saffle, and Wilson, 1998) provide some potentially attractive opportunities for CRPAQS in this regard. Located as shown on the map in Figure 2.1, several NEXRAD units are now operational in and around the CRPAQS study site. A prominent component of the National Weather Service modernization program, these radars have Doppler capabilities, with ranges extending to 450 km.

General features of the NEXRAD radars are summarized in Table 3.2. Although a variety of scanning sequences is possible, the units normally operate in one of two default scan modes, depending on whether precipitation exists within the radar domain, with the clear-air scan mode — which is of primary CRPAQS interest — being tuned to observe non-precipitation targets such as smoke, dust, pollen, birds, and insects. Data products returned from NEXRAD fall into four categories, which depend on the type of product and the degree of postprocessing desired. Most of these can be obtained either through the National Climate Data Center in Asheville NC or from official NEXRAD Information Dissemination Service vendors.

Tradeoffs between NEXRAD and transportable research radars involve several factors. Cost is certainly one of them, since temporary relocation and operation of the research radars, combined with downstream data processing, will involve significant expense. The beam-width of the NEXRADs, combined with their (at least current) inability to be synchronized in dual mode and their likely remoteness from local CRPAQS interest points, all contribute to relative lack of volumetric resolution and the inability to produce direct vector plots of three-dimensional wind fields. Moreover, even at minimum beam angles, the NEXRADS probe well above altitudes of CRPAQS interest, except for locations relatively close to the antennae. In addition the relatively long NEXRAD (C-band; 10 cm) wavelength makes them much more susceptible to ground clutter than their research counterparts, essentially precluding their usefulness for observing complex-terrain phenomena such as slope-flow circulations. The shorter wavelengths offered by the research radars (e.g. X-band; 3.2 cm and KA band; 0.87 cm for the NOAA radars) are sufficiently immune to ground clutter to enable their application in these areas. The NEXRADs, on the other hand, are in place. Moreover, the meteorological conditions of CRPAQS interest correspond to low-demand periods for the NEXRAD systems. This, combined with the generally positive and cooperative stance indicated by the NEXRAD administrative staff during telephone interviews for this report, suggests a flexibility and willingness to work cooperatively with CRPAQS scientists during field-study periods.

Table 3.2: Some General WSR-88D (NEXRAD) Features

Wavelength	10 cm
Gain	45 dB
Peak power	1 Mw
Beam width	1^{o}
Steerability	360^{o} azimuth 0.5^{o} - 19.5^{o} elevation
Range bin	250 m

As noted above chaff, tuned to resonant radar frequencies, is definitely attractive as a controlled, unambiguous radar target. Chaff releases, which are typically conducted by aircraft, will require close coordination with the FAA, the military, and the NWS, and will be essentially impossible in some locales such as FAA Control Zones, Federal Airways, and Control Areas. Because chaff particles settle with velocities of around 20 centimeters per second, periodic replenishment of chaff may be required, depending on the specific application.

3.7 Summary of Candidate Tracer Types

Table 3.3 summarizes the preceding material for the reader's rapid review while studying the tracer applications discussed in Sections 4 and 5. The "Relative Attractiveness" column in this table, a somewhat subjective set of judgements based on the discussion in this section, will be reflected in the more focused selection of tracers in later portions of this report.

Table 3.3: Summary of Candidate Tracer Types

Tracer Type	Advantages	Disadvantages	Relative Attractiveness to CRPAQS
Halogonated Organics	Sensitive; active and demonstrated technology; amenable to mobile sampling.	Point sampling; low temporal resolution, except for mobile samplers; relatively high cost encourages sparse network distributions.	Moderate to high
Additional Inert Gaseous Tracers	Sensitive; active and demonstrated technology for some gases	Point sampling; low temporal resolution, except for mobile samplers; relatively high cost encourages sparse network distributions;	Low to moderate
Fluorescent Particles (Bulk)	Inexpensive; depositing, thus emulates aerosol behavior.	Short-range applicability; size-distributed nature makes interpretation difficult.	Low
Single Fluorescent Particle Counting & Sizing	Inexpensive tracer; sensitive; provides aerosol size-discrimination as well as count.	Young technology; needs additional effort to build field-deployable system.	Moderate to high, if field system were immediately available. Otherwise low to moderate.
Fluorescent-Particle Lidar	Three-dimensional remote-sensing capabilities; low tracer cost.	Technology, while demonstrated, is apparently not currently at highly active level	Moderate to high, if field system were immediately available. Otherwise low to moderate.
Rare Earths	High sensitivity	High analytical costs; size-distributed nature makes interpretation difficult.	Low
Natural Radionuclides	Relatively uniform terrestrial source for radon (and thoron); radioactive decay provides opportunity to time transport processes. Mobile (aircraft) sampling as well as point sampling possible with current technology.	Experiment design often requires rather ideal conditions; data interpretation often involved and complex; some uncertainty regarding interpretive techniques in many cases.	Moderate

Table 3.3, Continued: Summary of Candidate Tracer Types

Tracer Type	Advantages	Disadvantages	Relative Attractiveness to CRPAQS
Research Doppler Radar	Three-dimensional remote-sensing capabilities; can provide three-dimensional wind fields in dual Doppler configuration; technology proven and available; can be applied in complex terrain situations.	Relatively expensive to deploy.	Moderate to high
NEXRAD Radar	Three-dimensional remote-sensing capabilities. Several units currently operating in CRPAQS domain; zero deployment cost.	Relatively coarse spatial coverage	Moderate to high

4. Screening Potential CRPAQS Tracer Applications: Objectives, Scientific Return, Costs, Design Considerations, and Probabilities of Success

This section presents a somewhat broad list of potential CRPAQS tracer applications, which is used as an initial screening forum to select a smaller subset that will be subjected to further scrutiny. A predominant theme throughout this section — one which will emerge as a final recommendation in Section 6 — is the assertion that any single tracer experiment conducted within CRPAQS should involve only a limited number of objectives, which are well posed and strongly focused: too large a profusion of objectives tends to blur experiment design, and thus limits the potential for obtaining meaningful and quantitative results. The application list presented below is somewhat modularized to accomplish this end. Having made this statement, however, it is important to note that some overlap exists between several of the "modules" discussed here, and that information from a tracer study designed to address a specific issue may be helpful in generating insights pertaining to others. The intent here is not to inhibit this beneficial overlap, but rather to encourage well-posed objectives and tight experiment design.

Questions to be used in evaluating the applications listed below are:

Question 1. Does the particular application address an essential information need to satisfy the CRPAQS objectives listed in Section 1? Assuming successful deployment, what is the potential scientific return?

Question 2. Do any of the tracer techniques listed in Section 3 lend themselves to this application? What is their probable cost-effectiveness and potential for successful deployment?

4.1 Evaluation of Large-Scale Flow Patterns (Meso-Beta — Meso-Alpha)

A number of perfluorocarbon tracer experiments within this category have been conducted previously in the San Joaquin Valley region and environs, with somewhat mixed results. Many of these have been designed to define footprints of ground-level tracer concentrations, and thus should fall into the "weak closure" category (Category 1) of Table 2.1. Primarily because of sparse sampler coverage and/or analytical sensitivity considerations, however, many can be more appropriately categorized into Category 2 (incomplete closure) or even Category 3 (qualitative hypothesis testing). While this information still can be considered useful in an "insight-generation" context, its value is diminished significantly under such conditions, causing some question as to the cost-effectiveness of past tracer applications of this type. There are several sub-classes of large-scale flow pattern applications, including:

- 4.1A Characterization of flows from the Bay Area into the San Joaquin and Sacramento Valleys, including flow divergences in the Sacramento Delta region.
- 4.1B Documentation of possible westward flows from the Valley areas through the Carquinez Strait region and into the Bay Area.
- 4.1C Documentation of major circulation patterns, such as the Fresno and Bakersfield eddies.
- 4.1D Documentation of flows from the Bay Area to the Livermore Valley and environs.
- 4.1E Characterization of land/sea return flows and their effects on the western boundary condition of the CRPAQS model domain.
- 4.1F Documentation of exit flows from the South SJV to the Mohave Desert.
- 4.1G Characterization of entrance Flows to the South SJV from the Los Angeles Area.
- 4.1H Differentiating transport/reaction behavior of pollutants (particularly NO_x) from high-elevation vs. low-elevation sources.

As indicated in Section 2.2, well-defined and quantitative tracer measurements of the above phenomena would be of substantial value to fulfilling several of the CRPAQS objectives, especially Objectives 1, 4, 7, and 8. Previously published reports combined with personal interview comments indicate that applications 4.1A, 4.1B, 4.1C, 4.1E, and 4.1H are especially important given the thrust of CRPAQS and the current state of knowledge.

With regard to general questions of technical feasibility and cost-effectiveness, the somewhat limited data applications — and high costs — associated with several past perfluorocarbon tracer applications is of some concern. Given the recent improvements in perfluorocarbon tracer technology noted above and armed with insights from past efforts, however, it is not implausible that future, more advanced studies could provide high-quality, quantitative information of the type desired. It is likely that augmentation of the perfluorocarbons with other tracer applications (e.g., radar or lidar) in key localities to provide additional three-dimensional information will be a desirable ingredient of any such study. Adaptation of a single-particle aerosol tracer component as a supporting function is also an option for consideration.

It is also essential that any tracer study within the above categories be closely coupled with modeling applications, both for evaluation of the models and for maximizing interpretation of the measured results. It seems obvious that any effort of this type will be a high-cost operation; in view of the noted relevance to

CRPAQS objectives, however, the potential return of tightly designed and well conducted studies of this type is quite high.

Applications 4.1E and 4.1H pose rather severe technical problems. Application 4.1E's focus on the western boundary condition of the model domain addresses an important CRPAQS question. Given the extended overwater location of the western boundary, however, it is difficult to conceive of any tracer experiment having reasonable cost that could supply the information required to resolve this issue in a meaningful manner. Application 4.1H, associated with high-elevation vs. surface NO_x emissions, also addresses an important aspect, but lacking comprehensive 3-dimensional observations aloft it is difficult to conceive of a definitive tracer-based measurement program for its effective resolution. The potential of advanced fixed-wing aircraft and balloon measurements (Kerrin, 1998) during future years may increase this feasibility; but in the CRPAQS time-frame the potential for such a tracer application appears doubtful. For these reasons applications 4.1E and 4.1H are given lower priorities based on technical feasibility; and of this group, only applications 4.1A, 4.1B, 4.1C will receive additional attention in Section 5, where selected applications are described in more detail.

4.2 Evaluation of Smaller-Scale Flow Patterns (Meso-Gamma — Meso-Beta)

4.2A Characterization of daytime slope-flow circulation patterns
4.2B Characterization of nocturnal drainage winds

Information pertaining to these two sub-topics directly addresses Objectives 1, 4, 7, and 8. Especially during warmer seasons, upslope winds result in pronounced circulation patterns adjacent to the valley walls, enhancing vertical motion and possibly leading to ventilation out of the valley environs. Although alternative explanations for inversion enhancement are possible (see Section 5.4, below), Alapaty and Seaman (1998) have suggested that subsidence aloft, as a consequence of the slope-wind return flow, results in sufficient compressional heating to reinforce the synoptic inversion, thus strengthening the meteorological barrier to pollutant escape. In any event, daytime, warm-season slope circulation phenomena are potentially strong determinants of pollution transport and pollution levels in the valley regions.

Nocturnal drainage flows bring air from aloft to the valley floor, and under stable nighttime conditions are not expected to add or remove appreciable amounts of pollution to/from the valley system. Thus this application is judged to be of lesser direct importance to CRPAQS objectives than is the daytime circulation phenomenon and will not be considered further in this report.

Although it apparently has not been used in this capacity to date, research Doppler radar, which generates three-dimensional wind fields, is an attractive possibility for use on slope-wind applications. Perfluorocarbon tracers and possibly FP lidar are also viable options.

4.3 Evaluation of Vertical Exchange in Surface Boundary Layers

From the noted program objectives, the meteorology of the San Joaquin Valley region under high $PM_{2.5}$ conditions, and the conceptual models set forth in the CRPAQS documentation, it is strongly evident that vertical transport through stable layers, especially under wintertime conditions, is an item of particular programmatic interest. Tracer applications for vertical-transport rate measurement have included a number of near-source plume studies as well as some inferrential examinations of tracer data at remote distances, although most of these have not dealt specifically with transport through stable-layer boundaries. A few tracer studies of rather special design have occurred, which have attempted to infer vertical transport on somewhat indirect bases. As a recent California example, scientists from SAI conducted a carefully implemented local tracer study, involving central releases of SF_6 with multiple concentration measurements on concentric sampling arrays, with concurrent detailed measurements of winds, turbulence, and other meteorological parameters (Carr, et al. 1998). This study produced a rich set of meteorological measurements for subsequent analysis; however, the application of the SF_6 measurements for vertical-transport rate analysis is not obvious nor is it described in the study's final documentation. Still other specialized studies have performed aircraft releases of tracer near to boundary-layer interfaces, with attempted measurement of the tracer on the adjacent sides of the boundaries (Ching and Alkezweeny 1986). Owing to measurement difficulties associated with tracer location as well as with interpretive complexities, the results of these studies have been marginal, at best.

As noted in Section 3.4 measurements of radon and its progeny have been deployed to measure vertical exchange rates, taking advantage of the timing features made possible by the decay-product lifetimes. Such studies usually have entailed vertical-profile measurements and have depended heavily on the assumption of one-dimensional, quasi-steady state conditions. Essentially such conditions demand a spatially uniform and temporally constant radon emission rate over the influence region of the measurements, with zero horizontal divergence of the radon and progeny fields aloft.

Other types of tracers do not seem particularly well suited to this application. Inert gaseous tracers present difficulties related to source configuration and associated interpretive uncertainties. Larger fluorescent particles and (especially) radar chaff have terminal fall velocities that are likely to be large compared to trans boundary-layer transport rates expected in the valley wintertime environment.

Tracer measurements within this class respond directly to CRPAQS Objectives 1, 4, and 7, and indirectly to Objective 8; thus they are highly relevant in the context of Question 1, above. From a technical feasibility standpoint it appears that radon and its progeny may be the most attractive tracers for this purpose, although local deviations from a one-dimensional steady state and possibly other interpretive uncertainties are of some concern. Because of terrain uniformity in the San Joaquin Valley basin and because of the low-wind meteorological conditions of interest, however, CRPAQS may offer a rather ideal circumstance for deployment of this technique. This aspect will be examined further in Section 5.3.

4.4 Aerosol Source Apportionment

Tracer-augmented source-apportionment studies typically involve colocated releases of unique tracers at specific source locations, with statistical evaluation of downwind tracer and pollution concentrations to determine relative contributions to pollution at the points of concentration measurement. The Mohave study, which applied perfluorocarbon tracers in this manner (Green and Tombach, 1997), is an important recent example of this type of investigation. As their name implies, source-apportionment techniques attempt to identify the sources responsible for pollution at chosen receptor sites, and are thus directly relevant to Objectives 1, 5 and 6 of Section 1, and are indirectly relevant to Objectives 2, 3, and 8.

Source-apportionment techniques are most applicable to situations where the tracer mimics both the source configuration and the atmospheric behavior of the pollutant(s) of interest. Depending on the statistical analysis being performed, relatively short-term measurements of the tracer as well as the pollutant concentrations may be required. Quite obviously, these requirements are met more easily for well-defined sources, such as point sources, and for primary, nonreactive pollutants.

In general, the CRPAQS does not conform well to either the source-definition or the primary-pollutant requirements noted above. Much of the aerosol in the CRPAQS region is secondary in nature, forming from condensation and/or reaction products of gaseous precursors. Moreover, configurations of both aerosol and aerosol-precursor sources are, in general, diffuse and complex leading to major problems in representing source configurations.

Source-apportionment studies fall under Category 4 in Table 1.1, and do not require "closure" in the sense used in this report. Consequently the demand for extensive sampler coverage is reduced substantially, leading to potential cost savings over the closure-based applications discussed in Section 4.1. Costs for the field deployment of tracer-based source-apportionment studies can be estimated on the basis of information given in that section.

In a conclusion to this section, tracer-augmented source-apportionment techniques are judged highly pertinent to several CRPAQS objectives, but rank low in technical feasibility.[4] Because of this, these methods will not be examined further in this report.

4.5 Evaluation of Aerosol Lifetimes and Deposition Rates

Past applications of tracers for measurement of aerosol lifetimes and deposition rates have generally fallen into two major categories, as follows:

1. release of an aerosol tracer designed to simulate the aerosol of primary interest, with subsequent measurement of airborne concentrations and/or deposition amounts at points downwind (usually a weak-closure class of study);

2. colocated release of an inert, nondepositing tracer with the aerosol of interest, with subsequent measurement of concentration ratios downwind (ratioing class of study).

4.5.1 Wet Deposition

Several of the aerosol tracers described in Section 3 could be deployed for wet-deposition studies. The meteorological conditions associated with high aerosol loadings in California do not involve precipitation, however, and thus wet deposition is of low interest to the CRPAQS. It is not relevant to any of the CRPAQS objectives, and will not be discussed further in this report.

4.5.2 Dry Deposition

Dry deposition is likely to be a primary, albeit slow, sink for aerosol within the study area during stagnation conditions, and thus is probably an important

[4] Note that this statement does not preclude CRPAQS application of non tracer-based source-apportionment techniques, which may be generate highly useful information for the study.

determinant of particulate-matter loadings during episodal conditions involving persistent high-pressure episodes. As such this subject is important to the fulfillment of CRPAQS Objectives 1, 3, 5, 7, and 8. Unfortunately, very few of the possible tracer studies lend themselves to providing definitive information on this subject. Ratioing studies are probably out of the question (at least for actual sources, see below), owing to the same confounding factors associated with diffuse source distributions and secondary particle generation that were discussed in Section 4.4. Simulant tracers have the significant problem of uncertainties regarding the tracer's ability to simulate the aerosol of interest. Moreover, studies involving bulk, heterodisperse tracer releases and aggregate measurements are strongly discouraged, owing to the strong relationship between particle size and deposition rate combined with the dominance of aerosol mass by the larger particles: These features typically combine to obscure much of the useful information desired from such efforts. Because of these features, it is probably most appropriate for the CRPAQS to avoid performing tracer-based aerosol-deposition studies and instead rely on the admittedly imperfect base of measurements available in the literature (e.g., Sehmel, 1980, 1984; Slinn, 1982).

There is one possible exception to this recommendation. This is the deployment of heterodisperse fluorescent-particle releases with downwind measurement of the resulting particle-size spectra, using the single-particle sizing and counting technique described in Section 3.3. This would allow measurement of deposition rates as a function of particle size, surmounting a majority of the problems noted in the previous paragraph. Moreover, and although the simulant's density, morphology, and possibly other physical properties cannot be expected to match that of the ambient "natural" aerosol, the fact that these are well characterized by the technique enhances the reliability of extrapolation of simulant properties to those of the aerosols of interest. This technique will be applied most appropriately in conjunction with a colocated release of a nondepositing tracer, allowing particle depletion to be measured by ratioing techniques.

4.6 Evaluation of Soil-Dust Resuspension

Resuspended dust is a significant contributor to local particle loadings, and evaluation of this phenomenon directly addresses CRPAQS Objectives 1, 2, 5, 7 and 8. During the past two decades a few studies have deployed tracers to assess rates of dust suspension from roadways, fields, and a variety of other soil surfaces (Sehmel 1983, 1984; Claiborn, et al. 1995). Although inert gaseous tracers have been deployed in some experiments, most short-term investigations of this type usually have involved application of tracer particles to the surfaces with subsequent measurement of airborne concentrations as functions of wind-speed, turbulence, traffic activity and/or other disturbing features. Longer-term studies have often involved deployment of solid tracer to a surface of well-defined geometry, with sequential measurement of adjacent soil samples at later times to examine translocation resulting from the resuspension-deposition process.

Often such studies have been plagued by uncertainties regarding particle size-distributions and particle-size dependence of the resuspension process, features that are complicated appreciably by the attachment of tracer to soil particles in largely unpredictable manners.

As a consequence of these and other factors, it is difficult to make a strong case for the application of tracers to measure resuspension processes in the CRPAQS context. Although the particle-size distributions of resuspended dust typically have tails that extend into the sub-2.5 micron region, dust is still not expected to be a major contributor to CRPAQS $PM_{2.5}$ episodes (cf. conceptual models for $PM_{2.5}$ episodes summarized in Section 2.2). According to the conceptual models for PM_{10} levels, namely

- Low winds, with nearby man-made dust sources, and
- High-wind dust suspension,

suspended dust certainly is a relevant contributor to coarse-particle episodes. Given the difficulties in tracer applications mentioned above, however, it seems much more productive and meaningful to examine suspension processes directly using measurements of the dust itself (possibly augmented by gaseous tracer measurements as described in the above-cited work by Claiborn and her co-workers), rather than some particulate tracer, which is likely to be a nonrepresentative and misleading simulant. For these reasons, tracer application for CRPQS measurement of dust-resuspension processes is given a low ranking in this survey.

4.7 Summary of Potential Tracer Applications

Table 4.1 summarizes the potential tracer applications discussed in this section, indicating judgements pertaining to tracer applicability, CRPAQS relevance, and technical feasibility by the sizes of the symbols in the respective matrix locations. From this and the above discussion, applications 4.1A, 4.1B, 4.1C, 4.1F, 4.2A, 4.3, and 4.5.2 appear to show sufficient merit to warrant further consideration. These are discussed individually in the following section.

Table 4.1: Screening Matrix for Potential CRPAQS Tracer Applications

Application Description	Possible Applicable Tracer Tecnologies	Relevance to CRPAQS Objectives	Technical Feasibility & Cost
4.1 Large-Scale Flow Patterns			
4.1A Document flows from the Bay Area into the San Joaquin and Sacramento Valleys; flow divergences in the Sacramento Delta region.	Perfluorocarbons ● Research Radar ● Fluorescent Lidar ● Fluorescent PC ● NEXRAD ?	1, 4, 7, 8 ■	Technically feasible, especially if perfluorocarbons used with research radar ♦ High cost
4.1B Document possible westward flows from the Valley areas into the Bay Area.	Perfluorocarbons ● Research Radar ● Fluorescent Lidar ● Fluorescent PC ● NEXRAD ?	1, 4, 7, 8 ■	Technically feasible ♦ Moderate to high cost
4.1C Documentation of major circulation patterns, such as the Fresno and Bakersfield eddies.	Perfluorocarbons ● Fluorescent Lidar ● Fluorescent PC ● NEXRAD ?	1, 4, 7, 8 ■	Technically feasible ♦ Moderate to high cost
4.1D Document flows from the Bay Area to Livermore	Perfluorocarbons ● Fluorescent Lidar ● NEXRAD ?	1, 4, 7, 8 ■	Technically feasible ♦ Moderate to high cost
4.1E Characterize land/sea return flows on the western model boundary	Perfluorocarbons ● NEXRAD ?	1, 4, 7, 8 ■	Infeasible ♦ Cost irrelevant
4.1F Document exit flows from the South SJV to the Mohave Desert	Perfluorocarbons ● Fluorescent Lidar ● NEXRAD ?	1, 4, 7, 8 ■	Technically feasible ♦ Moderate to high cost
4.1G Characterize entrance flows from the Los Angeles Area.	Perfluorocarbons ● Tracers of opp. ● Fluorescent Lidar ●	1, 4, 7, 8 ■	Technically feasible ♦ Moderate cost
4.1H Distinguish behavior of high- and low-elevation pollution releases	Perflurocarbons ●	1,3,5,6,7,8 ■	Infeasible ♦ Cost irrelevant

Table 4.1, Continued: Screening Matrix for Potential CRPAQS Tracer Applications

Application Description	Possible Applicable Tracer Tecnologies	Relevance to CRPAQS Objectives	Technical Feasibility & Cost
4.2 Smaller-Scale Flow Patterns			
4.2A Characterize daytime slope-flow circulation patterns	Perfluorocarbons ● Research radar ● Fluorescent lidar •	1, 4, 7, 8 ■	Technically feasible ♦ Moderate to high cost
4.2B Nocturnal drainage winds	Perfluorocarbons ● Research radar ● Fluorescent lidar •	1, 4, 7, 8 ■	Technically feasible ♦ Moderate to high cost
4.3 Evaluation of Vertical Exchange in Surface Boundary Layers	Radon ?	1, 4, 7 8 indirectly ■	Technically feasible (?) ♦ Moderate cost
4.4 Aerosol Source Apportionment		1, 5, 6 2, 3, 8 indirectly ■	Low technical feasibility ♦ Cost irrelevant
4.5 Evaluation of Aerosol Lifetimes and Deposition Rates			
4.5.1 Wet Deposition		Irrelevant to CRPAQS objectives	Feasibility and cost irrelevant
4.5.2 Dry Deposition	Fluorescent PC ● Bulk Fluor. Particles •	1, 3, 5, 7, 8 ■	Tracers not recommended, unless fluorescent PC is used; and then only for proof of principle purposes ♦
4.6 Evaluation of Soil-Dust Resuspension	Bulk Fluor. Particles •	1, 2, 5, 7, 8 ■	Low technical feasibility ♦ Cost irrelevant

5. Selected Tracer Applications

This section presents a somewhat more detailed discussion of the seven candidate tracer applications selected in Section 4. As was noted in Section 1, these discussions are not intended to compose detailed experiment designs, but rather to provide launch points for further pursuit by CRPAQS scientists should this be deemed advisable. In review, the selected applications are:

- 4.1A Characterization of flows from the Bay Area into the San Joaquin and Sacramento Valleys, including flow divergences in the Sacramento Delta region.
- 4.1B Documentation of possible westward flows from the Valley areas through the Carquinez Strait region and into the Bay Area.
- 4.1C Documentation of major circulation patterns, such as the Fresno and Bakersfield eddies.
- 4.1F Documentation of exit flows from the South SJV to the Mohave Desert.
- 4.2A Characterization of daytime slope-flow circulation patterns
- 4.3 Evaluation of Vertical Exchange in Surface Boundary Layers
- 4.5.2 Dry Deposition

These are discussed sequentially in the subsections immediately following.

5.1 Characterization of Flows from the Bay Area into the San Joaquin and Sacramento Valleys, Including Flow Divergences in the Sacramento Delta Region.[5]

This application is intended for completion within the Fall campaign during meteorological situations conducive to light-wind transport from the west under high-pressure synoptic conditions. It is intended, essentially as a partial-closure study in the sense of Section 1.3, and includes the following elements:

[5] Numerous, less comprehensive tracer studies have examined this effect. The reports of these studies should be reviewed carefully in the process of formulating the final design of this application.

Perfluorocarbon tracer release: Conduct a continuous, several-hour release at an elevated point (~100 m) west of the Carquinez Strait (Richmond-Rodeo locale). Consider sequential releases of multiple tracers as an option.

Ground-level tracer sampling: Locate an array of automated tracer samplers across the delta region and a limited north-south portion of the valley, ranging south to Modesto and north to Marysville. Maintain relatively tight sampler spacing within this limited spatial domain to guarantee good containment (closure) of the plume footprint over the study area. A small number of "sentinel" samplers may be placed at strategic, remote locations (e.g. Tehachapi) as well, to serve as indications (but not quantitative characterizations) of longer-range transport features.

Airborne tracer sampling: Deploy a light aircraft with onboard quasi-real time perfluorocarbon detection to provide a partial characterization of tracer transport and dispersion aloft. Do not constrain the aircraft to sample at a constant, pre-defined altitude; rather, design the flight strategy to maximize return of information aloft, especially regarding transport in the expected flow-divergence region.

Research Radar: Deploy a dual-Doppler system proximate to the main location of suspected flow divergence in the Delta region. Deploy a light aircraft with a continuous chaff cutter to maintain visible chaff concentrations throughout this region during the period of perfluorocarbon release.

NEXRAD: Post CRPAQS personnel to view NEXRAD scans at the two sites near Sacramento and the one at Hanford to detect larger-scale motions. Acquire Level-2 data tapes from these facilities for possible further analysis.

Possible additions/replacements: Consider deployment of FP lidar as an augmentation of the perfluorocarbon sampling system. This should be considered only upon confirmation of the state of readiness of a reliable and deployable system.

Data processing and model analysis: Consolidate the data obtained in this effort with profiler, sounder, and surface meteorological data taken as part of the Fall campaign. Perform high grid-resolution MM5 modeling to simulate local meteorology during tracer-release periods. Use the comparative results to verify/upgrade the model, analyze flow and divergence behavior, and to initiate an analysis of similar inflow events for climatological depiction.

Contact Persons:
Perfluorocarbon tracer release, sampling and analysis:
Russel Dietz, Brookhaven 516/344-3059

Doppler radar
Robert Kropfli, NOAA 303/497-6235
NEXRAD
Timothy Crum, NWS 405/366-6510
FP lidar
Edward Uthe, SRI 650/859-4667
Observational interpretation
Ted Smith, private consultant 626/795-2313
Modeling interpretation
Nelson Seaman, Penn State U. 814/863-3166

5.2 Documentation of possible westward flows from the Valley areas through the Carquinez Strait region and into the Bay Area.

This application is intended for completion within the Winter campaign during meteorological situations conducive to drainage transport under stable, low-wind, high particle-loading conditions. As with 5.1 above, it is intended as a partial-closure study in the sense of Section 1.3. In contrast to Application 5.1, however, this application deals with low-inversion, foggy conditions, and precludes (or at least discourages) the use of aircraft and radar observations, as well as other remote-sensing applications. As such, this application is confined to the following tracer deployments, which should be supported heavily by chemical and meteorological observations conducted during the wintertime intensive study.

Perfluorocarbon tracer release: Conduct continuous, simultaneous, 24-hour surface releases of two perfluorocarbon tracers, respectively located at two specified points in the Southern Sacramento Valley and the Northern San Joaquin Valley.

Ground-level tracer sampling: Locate an array of automated tracer samplers at low-elevation sites to the north and south of the Carquinez Strait, as well as strategic low-elevation points to the west.

Data processing and model analysis: Consolidate the data obtained in this effort with profiler, sounder, and surface meteorological data taken as part of the Winter campaign. Perform high grid-resolution MM5 modeling to simulate local meteorology during tracer-release periods. Use the comparative results to verify/upgrade the model and analyze valley drainage behavior.

Contact Persons:
Perfluorocarbon tracer release, sampling and analysis:
Russel Dietz, Brookhaven 516/344-3059

Observational interpretation

Ted Smith, private consultant 626/795-2313

Modeling interpretation

Nelson Seaman, Penn State U. 814/863-3166

5.3 Documentation of Major Circulation Patterns Such as the Fresno and Bakersfield Eddies, and Documentation of Exit Flows from the South SJV to the Mohave Desert

These two applications, which are intended for early in the Fall campaign period, will be discussed together here owing to their close relationship. These are intended, essentially, to be partial closure applications, in the sense of Section 1.3. Past meteorological analyses, tracer-based and otherwise, have suggested the existence of major eddy systems in the Fresno and Bakersfield areas during warm seasons running through September. Evidence indicates that during nocturnal conditions, at least, minimal pollution interchange occurs between these two eddies. One warm-weather scenario suggests the existence of quasi-independent nocturnal circulations aloft, which fumigate after sunrise and, aided by the upward movement of slope winds, deliver pollution to the Mohave over the Tehachapi range and, possibly, through Walker Pass.[6] The purpose of these applications is to document this behavior for subsequent modeling analysis and, ultimately, development of a climatology of these phenomena that is useful for pollution-management strategy development.

Perfluorocarbon tracer release: Perform simultaneous short-term evening releases of two perfluorocarbon tracers at strategically located, elevated points (~200 m) in the expected circulation patterns of the Fresno and Bakersfield eddies, respectively.

Ground-level tracer sampling: Locate a primary array of automated tracer samplers in a limited north-south portion of the valley ranging south to Tehachapi Pass and north to Madera. Maintain relatively tight sampler spacing within this limited spatial domain to guarantee good containment (closure) of the plume footprint over the study area. Locate a more sparsely spaced secondary sampler array at selected locations in the Mohave Desert to document transport to this area.

Airborne tracer sampling: Deploy a light aircraft with onboard quasi-real time perfluorocarbon detection to provide a partial characterization of tracer transport and dispersion aloft. Do not constrain the aircraft to sample at a constant, pre-defined altitude; rather, design the flight strategy to maximize return of

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[6] Much of the basis for this discussion originated from conversations and written input from Ted Smith of Pasadena, CA. The author expresses his appreciation for these helpful insights.

information aloft, especially with regard to cross-eddy tracer exchange, morning breakup of the tracer distributions, and subsequent transport to the Mohave.

NEXRAD: Post CRPAQS personnel to view NEXRAD scans at the Hanford and Edwards sites to detect larger-scale motions. Acquire Level-2 data tapes from these facilities for possible further analysis.

Data processing and model analysis: Consolidate the data obtained in this effort with profiler, sounder, and surface meteorological data taken as part of the Fall Intensive. Perform high grid-resolution MM5 modeling to simulate local meteorology during tracer-release periods. Use the comparative results to verify/upgrade the model, analyze circulation patterns, inversion-breakup, and exit-flow behavior, and to initiate an analysis of similar inflow events for climatological depiction.

Contact Persons:
Perfluorocarbon tracer release, sampling and analysis:
Russel Dietz, Brookhaven 516/344-3059
NEXRAD
Timothy Crum, NWS 405/366-6510
Observational interpretation
Ted Smith, private consultant 626/795-2313
Modeling interpretation
Nelson Seaman, Penn State U. 814/863-3166

5.4 Characterization of Daytime Slope-Flow Circulation Patterns

As noted in Section 4.2 there is some evidence that compressional heating from return-flow subsidence of the daytime slope-wind circulation may be sufficient to appreciably reinforce the stability of synoptic inversions in the valley regions. This is open to some question owing to the large width of the Valley and the smaller spatial scale of the circulation patterns; there are other plausible reasons why San Joaquin inversions may be particularly pronounced and persistent, such as topographically inhibited nocturnal cold-air drainage from the valley environs, and compressional heating by gravity waves on the lee side of the Diablo range during westerly flow conditions.[7] Regardless of the cause, it is important that the CRPAQS enhance the ability to understand and characterize the inversion-reinforcement and slope-wind phenomena. This is a partial - to - full closure application which is intended for operation early during the Fall campaign, and is designed to accomplish this goal. The location of the study should be on the western slope of the Sierra range, at a site that is conducive to aircraft and

[7] The author is indebted to David Whiteman of PNNL for his helpful discussions on this subject.

research-radar operations and maximizes the advantage of existing CRPAQS instrumentation.

Perfluorocarbon tracer release: Perform sequential, continuous (~1 hour) releases of multiple perfluorocarbon tracers at a selected location in the base inflow region of the slope-wind circulation.

Airborne tracer sampling: Deploy a light aircraft with onboard quasi-real time perfluorocarbon detection to provide a detailed characterization of tracer transport and dispersion aloft, especially the vertical extent of tracer transport and the horizontal and vertical extents of the return-flow pattern.

Research Radar: Deploy a single X- or KA-band radar at a location to remotely sense the travel and distribution of chaff deposited into the slope-wind circulation pattern. Deploy a light aircraft with a continuous chaff cutter to drop chaff into the slope-wind circulation from aloft during the period of perfluorocarbon tracer release.

Possible additions: Consider utilizing Doppler capability of research radar to derive radial velocity components, or even moving to a dual-Doppler configuration to derive velocity fields, if this is possible under the complex-terrain conditions associated with the application. Also consider the deployment of FP lidar as a possible replacement for the perfluorocarbon tracer system. It has been suggested also that any tracer study of this type be complemented by surface pollution samplers to provide additional information regarding impacts on Class I areas.

Data processing and model analysis: Consolidate the data obtained in this effort with profiler, sounder, and surface meteorological data taken as part of the Fall Intensive. Perform high grid-resolution MM5 modeling to simulate local meteorology during tracer-release periods. Use the comparative results to analyze the extent of compressional heating associated with the slope-wind circulation and its overall influence on inversions within the areal extent of the valleys.

Contact Persons:

Perfluorocarbon tracer release, sampling and analysis:
    Russel Dietz, Brookhaven 516/344-3059
Doppler radar
    Robert Kropfli, NOAA 303/497-6235
Observational interpretation and general field-study design
    David Whiteman, PNNL 626/795-2313
Modeling interpretation
    Nelson Seaman, Penn State U. 814/863-3166

5.5 Evaluation of Vertical Exchange in Surface Boundary Layers

As can be noted from the discussion in Section 4.3 this Winter campaign application has been selected for additional consideration more on the basis of its significance to CRPAQS than on the ensured technical feasibility of tracer techniques applied to its resolution. Transport estimation through vertically resolved radon measurements is essentially a full-closure class of study, but relies on some rather strong assumptions and idealized conditions regarding one-dimensionality and steady-state behavior. There appear to be several interpretive features requiring resolution in greater detail before any final CRPAQS decision to proceed on this front. In addition, this method must be supported by non-tracer instrumentation to resolve short-term phenomena, owing to the time-averaged nature of the measurement.

Vertical exchange during stable wintertime conditions is expected to be very slow with some diurnal variation, where it is likely that average transport occurs over long stagnant periods punctuated by infrequent breaking waves and other disturbing elements. Because the local radon concentrations will reflect averages of such behavior, it is essential that rapidly responding observations, such as a vertical-pointing lidar and sodar, be included to aid the interpretation. The Angiola site is specified here because, in addition to being well situated and possessing the 100 m tower, it will include sodar and other meteorological support instrumentation that will be helpful for data interpretation.

Radon-daughter measurements: Configure 3 to 5 radon-daughter monitors in a vertical array on the 100 m Angiola tower, extending from ground-level to the highest accessible location. Record data from these monitors throughout the period of the Winter Intensive. The desired degree of speciation of the daughter products provided by the monitors should be determined subsequent to more detailed scrutiny of the interpretive approach.

Lidar: Deploy a vertically pointed elastic-backscatter lidar at the Angiola site to document short-term influences on the inversion structure resulting from breaking waves or other infrequent, short-duration disturbances.

Data processing and model analysis: As noted above, a cursory review of the literature leaves some elements of data interpretation unclear regarding the radon profile measurements. This must be resolved, and the plan for data interpretation must be solidified before embarking on this application. Assuming success in this area, the results should be directly applicable to improving CRPAQS chemical-transport models.

Contact Persons:
Radon sampling and analysis
Vincent Negro, EML 212/620-3646
Observational and modeling interpretation
Sumner Barr, Los Alamos, NM 505/672-9410

5.6 Dry Deposition

This application was selected for further discussion because of the large significance of dry-deposition processes to the CRPAQS, and because of the high potential of the single-particle counting and sizing technology associated with fluorescent aerosol tracers. It can be deployed during the Long-Term, Fall, and/or Winter campaigns, and does not require extensive support instrumentation. It does, however, depend on the availability of at least one field-ready fluorescent-particle counter with demonstrated capabilities. Because some lead-time is necessary to achieve this state and also because of minimal prior experience in this field, CRPAQS should consider this application somewhat exploratory in nature, with a possibility of high return but also burdened with an appreciable contingency. It is best deployed as a ratioing class of application, in the sense of Section 1.3.

Fluorescent aerosol and perfluorocarbon release: Deploy a conventional aerosol generator to release a well-characterized dispersion of dye aerosol in precise proportion to a perfluorocarbon tracer. Although a wide spectrum of aerosol particles is acceptable, particle sizes should be maintained at sizes less than a few microns to minimize gravitational effects. Accurate knowledge of the initial size distribution, and its ratio to the perfluorocarbon release, are essential. The tracer-release location should be situated at a point where an extended downwind run over desired terrain features is expected.

FP counter and perfluorocarbon monitor: Deploy the fluorescent-particle counter and perfluorocarbon monitor in a van or other appropriate surface vehicle, suitable for intercepting the tracer plume at downwind points.

Data processing and interpretation: As noted above, data processing and interpretation is relatively simple since particle depletion, as a function of particle size, can be inferred directly from ratios of particle count to ambient perfluorocarbon concentration. Additional necessary data collection will include characterization of surface meteorology and land features for subsequent association with the corresponding deposition rates.

Contact Persons:

Perfluorocarbon tracer release, sampling and analysis:
Russel Dietz, Brookhaven 516/344-3059
Single FP instrumentation, sampling, and analysis
Lee Harrison, SUNY/Albany 518/437-8741

5.7 Summary Ranking of Tracer Applications

As noted above, the aim of this report is to provide basic information on tracer technology, as well as feasibility of the associated applications, so that the CRPAQS design team can evaluate options, establish priorities, and possibly select one or more applications for further consideration. Although this selection process is largely the purview of the design team, it is helpful to provide a basic ranking matrix to serve as an initiation point. Such a matrix is shown in Table 5.1, which ranks applications in descending order, based on the above discussions of presumed value, probability of success, and resource availability.

Table 5.1: Initial Ranking of Tracer Applications

Application	Value to CRPAQS	Success Probability	Technical Resource Availability	Cost
4.1A: Bay Area Inflow	8	8	8	High
4.1B Valley Outflows to Bay Area	7	8	8	Moderate
4.2A: Slope Flow Circulation	6	8	8	Moderate
4.1C/F S SJV Eddys/Exit Flows	6	7	8	High
4.3: Vertical Exchange	6	5	6	Moderate
4.5.2 Dry Deposition	5	5	5	Moderate

Based on the discussion in previous sections, the rankings here are largely self-explanatory. The "Value" category rankings for Applications 4.1C/F and 4.2A have been reduced somewhat owing to the fact that these phenomena are important only during warm seasons, and thus only during early periods of the Fall campaign.

6. Conclusions

This report was prepared using a variety of literature sources, including reviews and workshop reports on tracer technology (e.g., MATEX, 1984; Dietz, 1986; Barr, Clements, and Guthals, 1984), journal references, and field-study reports. In addition, interviews with a number of scientists conducted during preparation of this document (cf. *Acknowledgements*) were particularly helpful, and have been incorporated liberally into the text.

At the outset this work was intended to provide as lucid and sharply focused a product as possible, which would lead the reader to definitive conclusions regarding CRPAQS tracer deployment. Owing to the complex technical and logistical nature of tracer applications, however, this goal has been only partially realized: The selected candidate studies described in Section 5 still offer a variety of options to the reader, and others in the "long list" of Section 4 might be selected for further examination as well. Regardless of this, however, the report does provide a degree of focus that did not exist previously; in addition, it sets forth a reasonably objective screening procedure, which is likely to be helpful to anyone examining the subject further from a CRPAQS perspective. The initial ranking matrix presented in Table 5.1 is recommended as a starting point for this process.

A number of general recommendations can be made in support of, and in addition to, the material appearing in the earlier sections. These are:

1. *Future tracer studies should be designed to accommodate limited numbers of objectives, which are well-posed and realizable within the limitations of the measurement resource.* As noted in Section 4, manifold objectives have had a tendency to compromise the experiment designs of some past studies and thus limit the potential for obtaining quantitative results. The applications suggested in this report are purposely posed with limited numbers of objectives, in reflection of this recommendation.

2. *Long-distance tracer studies with widely extended sampler arrays, such as the 1990 SJVAQS perfluorocarbon study, should not be attempted in the CRPAQS.* This recommendation is somewhat a corollary of Recommendation 1, but is based on other considerations as well. Doubtless the 1990 perfluorocarbon study (Tracer Technologies, 1991) suffered from the sparse sampler layout necessitated by economic and logistical considerations. On the other hand, it did suffice to provide a number of valuable insights regarding transport phenomena (Lehman, et al. 1994), which should be useful indicators of behavior during the Fall campagin, at least early in the season. In view of this and also in view of the observational findings of Lehman and his co-workers, it is

evident that the 1990 data base has not yet been "mined" to the extent warranted by its potential usefulness. This leads to the following recommendation:

3. *The 1990 SJVAQS perfluorocarbon data base should be subjected to a comprehensive modeling analysis using MM5 and a suitable chemical-transport model, both to test the hindcasting ability of the models and to generate possible additional insights regarding summer and (possibly) fall transport behavior in the San Joaquin Valley and environs.*

4. *CRPAQS should examine the new NEXRAD radar array as an additional data resource for fulfilling programmatic objectives. This should involve making early arrangements with NEXRAD management for downstream interactions and formulating well-defined plans for data utilization, including defining data-acquisition pathways and data-processing protocols. Early plans for assigning and dedicating CRPAQS technical staff for this purpose should be made, to facilitate the downstream interaction during the field intensives.* Because of NEXRAD's ability to detect smoke and dust plumes, it should be considered as a potentially valuable resource, even if chaff tracers are not deployed in the CRPAQS. Since this is a new and largely untested resource (note question marks associated with NEXRAD entries on Table 4.1) extensive prior CRPAQS investigation will be required to explore this possibility.

Finally,

5. The CRPAQS should maintain cognizance of other field programs involving tracer measurement of atmospheric transport, which are currently in late design stages, are likely to field measurement campaigns prior to 2000, and have the potential for generating new information regarding tracer deployment that is of potential value to this program. Studies that may be of particular significance in this regard include:

• The Breton Air Monitoring Program (BAMP). Gulf of Mexico and Alabama, Louisiana, and Mississippi) Contact: Russel Dietz, Brookhaven 516/344-3059.

• The DOE Vertical Transport Program. Contact: Chris Doran, PNNL 509/372-6149.

• Project BRAVO. Big Bend area of Texas and Northern Mexico. Contact: Marc Pitchford, NOAA, 702/895-0432.

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