Allocation of terrestrial carbon sources using $^{14}CO_2$; measurement and modeling

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Abstract:

The radiocarbon content of whole air provides a theoretically ideal and now observationally proven tracer for recently-added fossil fuel derived CO_2 in the atmosphere (C_{ff}). Over large industrialized land areas, determination of C_{ff} also constrains the change in CO_2 due to uptake and release by the terrestrial biosphere. Here we review the development of a $\Delta^{14}CO_2$ measurement program and its implementation within the NOAA Global Monitoring Division's air sampling network, using the US as an example. The $\Delta^{14}CO_2$ measurement repeatability is evaluated based on surveillance cylinders of whole air and equates to a C_{ff} detection limit of ≤0.9 ppm based on measurement uncertainties alone. We also attempt to quantify additional sources of uncertainty arising from non-fossil terms in the atmospheric $^{14}CO_2$ budget and from uncertainties in the composition of "background" air against which C_{ff} enhancements occur. As an example of how we apply the measurements, we present estimates of the boundary layer enhancements of C_{ff} and C_{bio} using observations obtained from vertical airborne sampling profiles off of the northeastern US. We also present an updated time series of measurements from NOAA GMD's Niwot Ridge site at 3475 m asl in Colorado in order to characterize recent $\Delta^{14}CO_2$ variability in the well-mixed free troposphere.

1. Introduction:

The small radiocarbon fraction of total CO_2 has proven to be an ideal tracer for its fossil fuel derived component [c.f. *Levin et al.*, 2003; *Turnbull et al.*, 2006; *Graven et al.,* 2009; *Miller et al.*, 2012]. Unlike all other contributions to the atmospheric CO_2 budget, the fossil fuel component is devoid of radiocarbon, so that temporal and spatial gradients in recently added fossil fuel CO_2 can be readily identified as radiocarbon gradients provided there is adequate precision in the measurements. Over large industrialized land areas such as Eurasia and North America, the use of ^{14}C to isolate the contribution of recently added fossil fuel to the observed CO_2 mole fraction (C_{ff}) also quantifies (by difference) the change in atmospheric CO_2 due to uptake and release by the terrestrial biosphere (C_{bio}). Here we review the development of a precise $^{14}CO_2$ processing and measurement capability in discrete ~2 L (STP) samples of whole air, suitable for implementation within existing flask sampling networks around the world. Our development effort was undertaken as a collaboration between the University of Colorado, the Global Monitoring Division (GMD) of NOAA's Earth System Research Laboratory, and the Keck AMS Facility at the University of California at Irvine.

Most of our measurements occur within NOAA GMD's global air sampling network, which includes flask sampling at the surface, in aircraft profiles, and from tall towers [http://www.esrl.noaa.gov/gmd/dv/iadv/]. Paired ^{14}C and trace gas measurements expanded into a growing network of tall towers around the US between 2009 and 2010, facilitated in part by a new measurement collaboration with the Center for Accelerator Mass Spectrometry at Lawrence Livermore National Laboratory.

We begin by outlining the atmospheric budgets of CO_2 and $\Delta^{14}CO_2$ and an analytical framework for C_{ff} detection using paired CO_2 and $\Delta^{14}CO_2$ observations. We then document the $\Delta^{14}CO_2$ measurement repeatability as ascertained in surveillance cylinders of whole air and give an example of how authentic field measurements have been used to partition boundary layer enhancements of CO_2 into fossil fuel and terrestrial biospheric components. Finally, we describe uncertainties in our C_{ff} detection

methods arising from non-fossil terms in the isotopic budget and from incomplete representation of "background" air against which enhancements are determined.

2. C_{ff} detection using $^{14}CO_2$:

The value of radiocarbon measurements with regard to C_{ff} detection arises from the fact that ancient carbon comprising fossil fuels is devoid of ^{14}C due to radio-decay while the ^{14}C activity of the atmosphere and living biosphere is maintained at relatively high (i.e. "modern") levels as a result of ongoing ^{14}C production. Thus, CO_2 sourced from the combustion of fossil fuels is ^{14}C-free whereas all other sources of CO_2 to the atmosphere have isotopic signatures that are close to that of the atmosphere itself. In simplified form, $\Delta^{14}C \approx [(^{14}C/C)_{sample}/(^{14}C/C)_{standard}-1]1000‰$, but with corrections for mass-dependent fractionation (from $\delta^{13}C$ measurement) and small amounts of radioactive decay between the times of sampling and measurement (see *Stuiver and Pollach* [1977] for full expression). Therefore, the $\Delta^{14}CO_2$ of fossil fuel CO_2 = -1000 ‰. In contrast, the measured value of the recent, well-mixed atmosphere is about +50 ‰ [*Turnbull et al.,* 2007; *Levin et al.,* 2008; *Graven et al.,* 2012]. By mass balance, the addition of 1 ppm of fossil CO_2 to an atmospheric CO_2 burden of 390 ppm will produce a $\Delta^{14}C$ depletion of ~2.7 ‰ (i.e. (-1000 ‰ - 50 ‰)/390 ppm).

We describe the global atmospheric budgets for CO_2 and its ^{14}C:C ratio (expressed in the Δ notation) below in equations 1a and b;

$$\frac{dC_{atm}}{dt} = F_{bio} + F_{oce} + F_{fos} \qquad (1a)$$

$$C_{atm}\frac{d\Delta_{atm}}{dt} = (\Delta_{fos} - \Delta_{atm})F_{fos} + \Delta_{ocedis}F_{ocedis} + \Delta_{biodis}F_{biodis} + isoF_{nuc} + isoF_{cosmo} \qquad (1b).$$

For the CO_2 budget (equ. 1a), C_{atm} refers to the atmospheric mole fraction of CO_2, F_{bio} represents the net terrestrial biosphere-atmosphere flux, F_{oce} is the net ocean-atmosphere flux, and F_{fos} is the flux from fossil fuel combustion. For the isotopic budget

(equ. 1b), net isotopic exchange terms can be neglected since the Δ notation includes a $\delta^{13}C$ correction that accounts for mass dependent fractionation. The subscripts "ocedis" and "biodis" denote the ocean-atmosphere and biosphere-atmosphere isotopic disequilibria and associated gross (one-way) mass fluxes. Isotopic disequilibrium refers to the difference between isotopic signatures of carbon leaving and entering a reservoir. In the terrestrial case, disequilibrium results from the respiration of ^{14}C-enriched CO_2 assimilated, on average, a decade or two ago when the atmospheric $\Delta^{14}C$ was much higher as a result of atmospheric nuclear weapons testing. In the oceanic case it results from the reemergence of aged ^{14}C-depleted CO_2 at the surface, carried by waters which have been out of contact with the atmosphere long enough for radioactive decay to become significant. The subscript "nuc" refers to the flux of $^{14}CO_2$ from nuclear reactors, and "cosmo" to the cosmogenic production of ^{14}C. These last terms are pure ^{14}C fluxes and as such don't have isotopic signatures and are represented only as "*iso*"-fluxes. If the non-fossil contributions to the tropospheric ^{14}C budget are either small or relatively uniform in space, then the mass balance sensitivity and the ^{14}C measurement precision will closely approximate the actual C_{ff} detection capability.

To illustrate this, we show in ***Figure 1*** a map of wintertime fossil fuel-derived CO_2 and (total) $\Delta^{14}CO_2$ within the planetary boundary layer (PBL) over North America as represented in the TM5 transport model [*Krol et al.*, 2005; Miller *et al.*, 2012]. For $^{14}CO_2$, all terms in eq. 1b are represented in the model, except the nuclear term [*c.f., Graven and Gruber*, 2011]. The color scales depicting $\Delta^{14}CO_2$ and fossil fuel CO_2 distributions correspond to the expected mass balance sensitivity of -2.7 ‰/ppm. Thus, the similar colors and patterns in ***Figure 1*** indicate that, over North America, the ^{14}C gradients are controlled largely by the presence of fossil fuel CO_2. The remaining small differences are due primarily to small atmospheric gradients imposed by the terrestrial disequilibrium flux of ^{14}C ($\Delta_{biodis}F_{biodis}$ in eq. 1b). This contribution can be quantified and applied as a small correction in the fossil fuel CO_2 detection algorithm, as discussed below. ^{14}C emissions from nuclear power generation [*Graven and Gruber*, 2011], which are neglected in this simulation due to large relative uncertainty, may produce near-surface

signals averaging 1 to 2 ‰ in the densely populated northeastern US (see *Section 5* for a discussion of related C_{ff} detection uncertainties). The cosmogenic production and ocean disequilibrium terms do not result in significant gradients of ^{14}C over the US, although they are also specified in the model. Isotopic budget terms used in the TM5 simulations have been estimated following methods outlined in Turnbull *et al.* [2009] and at CarbonTracker [http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/documentation.html].

In order to quantify the fossil fuel CO_2 signal from measurements, we follow Levin *et al.* [2003] in considering observations of both CO_2 and $\Delta^{14}C$ to be the sum of background values for each tracer plus any fossil fuel and biospheric contributions;

$$C_{obs} = C_{bg} + C_{ff} + C_{bio} \quad (2a)$$

$$\Delta_{obs}C_{obs} = \Delta_{bg}C_{bg} + \Delta_{ff}C_{ff} + \Delta_{bio}C_{bio} \quad (2b).$$

As in Turnbull *et al.* [2006], we divide C_{bio} into photosynthetic and respiratory terms C_{photo} and C_{resp}, respectively. Expanding and combining eqs. 2a and b, and setting Δ_{photo} equal to Δ_{bg} (which will be the same as a result of ^{13}C:^{12}C normalization), we obtain

$$C_{ff} = \frac{C_{obs}(\Delta_{obs} - \Delta_{bg})}{\Delta_{ff} - \Delta_{bg}} - \frac{C_{resp}(\Delta_{resp} - \Delta_{bg})}{\Delta_{ff} - \Delta_{bg}} \quad (2c).$$

In eq. 2c, all of the quantities in the first term on the right-hand-side are either known *a priori* or can be measured, and the second term, which we call C_{corr_resp}, is a correction to C_{ff}, which accounts for the disequilibrium contribution of ^{14}C from heterotrophic respiration. The recent isotopic disequilibrium is approximately [*Ciais et al.*, 1999] the difference between present day atmospheric $\Delta^{14}C$ and that from a decade or so earlier, reflecting the mean residence time of carbon in the terrestrial biosphere. C_{corr_resp}, which always acts to lower C_{ff}, was estimated previously as 0.4 - 0.8 ppm in summer and 0.2 – 0.3 ppm in winter, based on estimates of the seasonally varying

heterotrophic respiration flux and PBL height and the mean terrestrial biosphere isotopic disequilibrium [*Turnbull et al.*, 2009; *Turnbull et al.*, 2006]. Miller *et al.* [2012] recently introduced a method for estimating the C_{corr_resp} correction term on a sample-by-sample basis which we will discuss below in *Section 5*.

Having estimated C_{corr_resp}, the C_{ff} enhancement relative to background is calculated from eq. 2c. Equation 2a can then be applied to isolate C_{bio}, which is the biological enhancement or depletion of CO_2 relative to background.

3. $^{14}CO_2$ Measurement precision and C_{ff} detection limit:

Taking the simulated distribution of $\Delta^{14}CO_2$ in the near surface over the US (for a week in January, 2006) in ***Figure 1a*** as an example, we may expect continental scale gradients within the PBL of up to 10-15‰ in winter, suggesting that a $\Delta^{14}CO_2$ measurement precision of better than 2‰ would be needed in order to resolve synoptic gradients with confidence (i.e., with a signal to noise ratio of $\geq$ 5:1). Time dependent $\Delta^{14}CO_2$ variability within the PBL at observing sites in the US, Europe and E. Asia may reach 10 to 20 ‰ [*c.f., Levin et al.*, 2003; *Turnbull et al.*, 2011; *Miller et al.*, 2012] but is often much less, depending on transport and proximity of observing sites to major fossil CO_2 combustion sources. We also see from eqs. 2b and c that quantification of fossil fuel CO_2 enhancements from observations requires measurements in both "background" and "observed" air for both the CO_2 mole fraction and its ^{14}C activity. Propagation of measurement uncertainties through eq. 2c suggests that in order to obtain a C_{ff} detection limit of ≤1 ppm (1σ), the necessary $\Delta^{14}CO_2$ measurement precision will be ≤2 ‰ (1σ), with the C_{ff} detection limit determined almost entirely by the much larger relative uncertainty in the ^{14}C measurement precision (i.e., ~1/20 for ^{14}C vs. ~1/4000 for CO_2 if the CO_2 measurement precision is ±0.1 ppm, [*Conway et al.*, 1994]). For long-term monitoring programs, the relevant metric of precision will be the long-term ^{14}C measurement repeatability (defined by *JCGM* [2008]) in appropriate control materials rather than

178 the single-sample measurement precision (sometimes also called the "external
179 precision" when using AMS).
180 At the University of Colorado, the long-term measurement repeatability is
181 evaluated based on repeat extraction and measurement of whole air from control
182 cylinders. From 2003 to 2009, we used for this purpose a single cylinder of whole air
183 filled at NOAA's Niwot Ridge, CO site (NWR, at 3475 m asl) in Sept. 2002, which we
184 refer to as "NWTstd" [*Turnbull et al.*, 2007]. In 2009, by which time NWTstd was near
185 exhaustion, we introduced two new control cylinders, "NWT3" and "NWT4". Both
186 were filled at NWR in Feb. 2009, but a small amount of ^{14}C-free CO_2 was added to
187 NWT4 to lower the activity with respect to ambient values. Presently we measure 3
188 extraction aliquots each from NWT3 and NWT4 in each AMS measurement wheel.
189 Individual measurement wheels are typically comprised of 8 primary measurement
190 standards (NBS Oxalic Acid I, Ox-I), 1 secondary standard (NBS Oxalic Acid II, Ox-II), 1
191 process blank (^{14}C-free CO_2 in air from cylinders), 6 controls (NWT3 and 4), and 24
192 authentic samples. Extraction, graphitization, measurement and normalization follow
193 methods described in *Turnbull et al.* [2007]. Extraction occurs on either a manual or an
194 automated extraction line ("CRex", as described in *Turnbull et al.,* [2010]) and samples
195 from individual observing sites are always extracted on the same line.
196 In ***Figure 2*** we show individual measurement values, means and raw (un-
197 normalized) 1σ repeatabilities for both NWT3 and 4 by order of either measurement
198 or extraction, from September 2009 to May 2012. There is no statistically significant
199 difference between results for the two extraction lines. Temporal patterns of the
200 measured value by measurement order suggest some systematic variances result from
201 either graphitization or measurement conditions (graphitization and measurement
202 occur nearly simultaneously). This may be expected, despite the large number of
203 accompanying primary measurement standards, since, in our experience, the within-
204 wheel agreement of controls from gas cylinders is generally better than that for Ox-I
205 [*Lehman et al.*, 2010].

206 Long-term repeatabilities for NWT3 and 4 of 1.6 – 1.7 ‰ (***Fig. 2***) are slightly
207 better than but comparable to the values of 1.8 - 1.9‰ previously reported for
208 NWTstd from 2003-2009 [*Turnbull et al.,* 2007*; Lehman et al.,* 2010]. For authentic
209 samples, we continue to report a 1σ measurement repeatability of ±1.8 ‰ or the
210 single sample measurement precision, whichever is larger [*Turnbull et al.*, 2007].
211 Propagation of uncertainties of ±1.8 ‰ for $\Delta^{14}CO_2$ and of ±0.1 ppm for CO_2 through
212 eq. 2c suggests a long-term detection limit for C_{ff} of 0.9 ppm, based on measurement
213 uncertainties alone. This will also characterize the uncertainty in C_{bio} determined
214 according to eq. 2a.
215

216 ## 4. A Northeast US example:
217
218 As an example of how we apply actual field measurements, we show in ***Figure***
219 ***3*** the separation of boundary layer enhancements of CO_2 into their fossil fuel and
220 biological components, using ~6 years of airborne observations at CMA and NHA (from
221 *Miller et al.*, [2012]), in a region of significant emissions and air outflow in the
222 northeast US (see ***Fig. 1*** for site locations). In this analysis, boundary layer samples
223 from ~300 m asl are treated as "*obs*", and "*bg*" is from ~4000 m asl within the well-
224 mixed free troposphere sampled in the same vertical profile. This treatment of the
225 observations effectively assumes that the overlying free troposphere is the source of
226 air into which new emissions are added in the PBL. The presentation in ***Figure 3*** differs
227 from that in *Miller et al.* [2012], in that here we decompose only the observations
228 from ~300 m asl, as opposed to those from ~300 m asl and from 2000-2400 m asl,
229 since the latter are often above the PBL.
230 We see that C_{ff} is present within the PBL throughout the year, with ~80% of the
231 observations in the range of 0 - 7 ppm. Occasional instances of negative C_{ff}, which are
232 non-physical, are explained almost entirely by the 1σ $\Delta^{14}CO_2$ measurement
233 uncertainty, and to a far lesser extent by the failure of our simplified assumptions
234 regarding the source of "backround" air [*Miller et al.*, 2012]. Accounting explicitly for

C_{ff} using ^{14}C also sharpens our view of the biospheric CO_2 signal, which differs significantly from the raw boundary layer enhancement or depletion of CO_2 (i.e., the difference between green and black lines in ***Fig. 3***). Also noteworthy is the large presence of respiratory CO_2 in the boundary layer in winter, when, on average, the biological emissions account for ~60% of the total PBL CO_2 enhancement. This suggests that reliable attribution of sources using ground- or satellite- based, CO_2-only observing strategies will not be possible without additional information, even over urban and industrial areas in winter.

5. Uncertainties from correction terms:

In the analysis above, the correction term C_{corr_resp} (the 2nd term in eq. 2c) was calculated explicitly for each sample according to methods detailed in *Miller et al.* [2012]. We use these estimates here as a quantitative basis for assessing related uncertainties in C_{ff}. The heterotrophic respiration flux as a function of time since assimilation was estimated monthly at a resolution of 1° x 1° using pulse response functions from the CASA biosphere model [*Thompson and Randerson*, 1999] and then convolved with the atmospheric $\Delta^{14}CO_2$ history [c.f., *Levin and Kromer,* 2004] to obtain $F_{biodis}\Delta_{bios}$ (eq. 1b). This was applied to the individual observations (as the numerator in C_{corr_resp}) using spatial sensitivities to surface emissions obtained for each sample from a Lagrangian transport model (FLEXPART, [*Stohl et al.*, 2005]). The individual corrections along with their monthly mean and standard deviation for all observations at CMA are given in ***Figure 4***. As expected, the estimates for the ~300 m asl observations within the PBL show a larger sensitivity to the surface emissions than the samples from 2000 – 2400 m asl. The monthly average estimates for the PBL of ~0.7 ppm in summer and ~0.3 ppm in winter are comparable to previous estimates of the seasonal mean corrections [*Turnbull et al.*, 2006; *Turnbull et al.*, 2009]. The likely uncertainty associated with applying seasonal mean corrections as opposed to explicitly determined sample-by-sample corrections is indicated by the monthly 1σ standard deviations, which are up to

~0.4 ppm in summer and ~0.1 ppm in winter. Associated corrections and uncertainties will likely be smaller for regions dominated by relatively dry Mediterranean climates, compared to those for the humid continental eastern US that apply here. The absolute uncertainty is difficult to determine, but will stem primarily from the accuracy of the biospheric residence times in the CASA biosphere model.

As stated earlier, we have neglected the influence of $^{14}CO_2$ emissions from the nuclear power sector in the TM5 simulations and in eq. 2c, which is used to determine C_{ff} from observations. Using gridded estimates of the nuclear emissions from *Graven and Gruber* [2011] we calculate possible corrections for all observations at CMA using the same method as above. Like C_{corr_resp}, the term C_{corr_nuc} will always act to raise C_{ff}, since both are surface sources of $^{14}CO_2$ that mask the isotopic dilution associated with surface emissions of fossil fuel derived CO_2. As shown in ***Figure 5***, estimated C_{corr_nuc} shows little seasonal variation and is considerably larger in the near surface than at 2000-2400 m asl. Monthly means range from 0.25 – 0.9 ppm for the PBL, with monthly 1σ standard deviations of 0.15 - 1.0 ppm. Estimates of $C_{corr\text{-}nuc}$ and its monthly standard deviation for all observations at NHA (not shown) are approximately 50% lower. The annual mean signals agree within errors with those for the northeast US obtained by *Graven and Gruber* [2011] (using an Eulerian transport model) for their central estimates of nuclear emissions. The predicted gradient in nuclear signals between CMA and NHA is due to the distribution of reactors (there are substantially fewer in northern New England than in the large metropolitan areas to the south and west [http://www.nrc.gov/reactors/operating/map-power-reactors.html]) and the approximate e-folding length scale for sensitivity to emissions influencing our 300 m asl observations at CMA and NHA of 150 – 300 km (from FLEXPART).

If the central emissions estimates of *Graven and Gruber* [2011] closely approximate the actual power plant emissions of $^{14}CO_2$, then we may conclude that likely maximum uncertainties in C_{ff} due to variability in C_{corr_nuc} are in the range of 0.1 to 1.0 ppm for the eastern US (based on estimates for CMA, influenced by a region in which reactors are relatively common). The potential low bias in C_{ff} that arises from

neglecting C_{corr_nuc} altogether has about the same range as the uncertainty (e.g., range of monthly means compared to range of monthly standard deviations in ***Fig. 5***). It is important to note, however, that the $^{14}CO_2$ emissions estimates are characterized by large uncertainties, which *Graven and Gruber* [2011] represent as 70% confidence intervals of the distributions of total ^{14}C emissions for different reactor types. In the US, all reactors are either pressurized- or boiling- water reactors, both of which emit ^{14}C as $^{14}CO_2$, although emissions from the more common pressurized water reactors (PWRs) are primarily in the form of methane. Estimates of the fraction of ^{14}C emitted as $^{14}CO_2$ for PWRs range from 5 – 25% [*IAEA*, 2004], which suggests substantial uncertainty in overall nuclear $^{14}CO_2$ emissions in the US. Estimates of the influence on C_{ff} for the lower limit of the 70% confidence interval for nuclear emissions are ≤ 0.25 ppm for the US [*Graven and Gruber*, 2011]. The influence associated with the upper limit of emissions estimates for the US is large enough to produce frequent and sustained reversals in the vertical ^{14}C gradient which are not observed (i.e., producing negative C_{ff} well in excess of that in ***Fig. 3***). The distributions used to obtain 70% confidence limits may, in fact, contain a high bias, as the fraction of ^{14}C emitted as $^{14}CO_2$ by PWRs was specified as 25%, which is at the upper end of independent estimates.

Regardless of the uncertainties, the nuclear signal in much of the central and western US will be negligible (with the possible exception of areas near 4 reactors in southern California and 3 in Arizona). In parts of Europe, the necessary corrections and uncertainties will be substantially larger due to the large number of reactors and reactor types, and the presence of fuel reprocessing plants [*Graven and Gruber* 2011; *Levin et al.,* 2003]. For ^{14}C-based C_{ff} observing programs in Europe and Japan, and very near reactor sites in the US, additional monitoring of nuclear $^{14}CO_2$ emissions may be needed [*Batler*, 2012].

6. Uncertainties from specification of "background":

Determination of the enhancement of fossil fuel derived CO_2 directly from observations requires measurements and/or estimates of both "observed" and

323 "background" air (i.e., eqs. 2a-c). In the case of the NE US example given above (*Section*
324 *4*), measurements from the overlying free troposphere sampled in the same vertical
325 profile as the PBL "observation" were used to represent "background". However, if air
326 refreshing the PBL (into which new emissions are then added) originates from locations
327 with a different CO_2 mole fraction and/or a different isotopic composition than the
328 contemporaneously overlying free troposphere, the estimated PBL enhancement would
329 be subject to error over and above that associated with measurement uncertainty
330 alone. In the case of seasonally persistent horizontal shear (in which upper and lower
331 level air originate from different locations), such errors would be present as an
332 undetected seasonal bias in C_{ff}.

333 Sustained meridional gradients of $\Delta^{14}CO_2$ in the free troposphere over northern
334 hemisphere continents are expected as a result of the changing balance of isotopic
335 budget terms (i.e., those in eq. 1b) with latitude [*Randerson et al.*, 2002; *Levin et al.*,
336 2010; *Graven et al.*, 2012], while strong upper level westerly winds will tend to suppress
337 zonal free troposphere $\Delta^{14}CO_2$ gradients in middle latitudes. For the period 2005-2007
338 (the most recent available observations), Graven *et al.* [2012] document a mid-latitude
339 minimum in average $\Delta^{14}CO_2$ in the Northern Hemisphere of ~3±1 per mil with respect to
340 observations at 15°N and of ~1±0.5 per mil with respect to 70 °N. These are comparable
341 to mean Northern Hemisphere gradients observed by Levin *et al.* [2010] from 1994-
342 1997, but in both studies the NH gradient is characterized by only 4 observing sites.

343 For the NE US example, *Miller et al.* [2012] provide an analysis of possible bias
344 resulting from seasonal shear along the east coast based on simulated $\Delta^{14}CO_2$
345 distributions over North America, since the actual recent $\Delta^{14}CO_2$ distribution remains
346 poorly constrained. Back-trajectories calculated by FLEXPART for CMA in summer show
347 that the high altitude samples originate further north by ~15˚ than do those for the
348 lower troposphere three days prior to sampling. Sampling the TM5 $\Delta^{14}C$ output using
349 the end points of 7 day back trajectories indicates that the trajectories for upper level
350 samples intersect simulated free troposphere $\Delta^{14}C$ values higher than those for the
351 lower level samples by ~1.6 ‰, equating to a possible seasonal C_{ff} bias of ~ +0.5 ppm

352 C_{ff}. During winter, the difference between upper and lower level endpoints is ~3˚ of
353 latitude three days prior to sampling, suggesting that the contemporaneously overlying
354 wintertime free troposphere provides a relatively reliable estimate of background.
355 We can evaluate the mean and variability of the recent zonal free troposphere
356 $\Delta^{14}CO_2$ gradient over North America by comparing measurement results from NWR (at
357 3475 m asl, 40.05° N) and upper level (~4000 m asl) samples from CMA (at 38.83 °N) and
358 NHA (at 42.95 °N) for the same time periods. In ***Figure 6a*** we show an updated $\Delta^{14}CO_2$
359 time series for NWR for the period 2004-2011 (extending results in *Turnbull et al.*,
360 [2007]), with observations associated with >2-sigma outliers in corresponding
361 measurements of CO removed in order reduce the influence of local pollution events.
362 Also given is a combined polynomial and harmonic fit to the observations that is
363 frequently used to isolate the seasonal signal in atmospheric CO_2 observations [*Thoning*
364 *et al.*, 1989]. The one-sigma deviation of the individual NWR observations from the fit is
365 ±1.9 ‰, similar to the long-term measurement uncertainty.
366 In ***Figure 6b*** we compare the individual ~4000 m asl observations from CMA and
367 NHA (from *Miller et al.*, [2012]) to the NWR fitted curve. The mean deviation of the NHA
368 observations from the NWR fit is +0.07 per mil, with a one-sigma deviation from the
369 NWR fit of ±2.2‰, comparable to that for the NWR observations themselves. For CMA,
370 the mean and one-sigma standard deviations with respect to the NWR fit are +0.61 ‰
371 and ±2.6 ‰, respectively. Positive deviations of the individual observations with respect
372 to NWR occur at both eastern sites in summer (Fig. 4b), possibly due to weaker
373 summertime mean westerlies and increased zonal flow. The shared variability at
374 different sites sampling the relatively well-mixed free troposphere likely arises from
375 changes in vertical transport in addition to seasonal changes in sources [*Turnbull et al.*,
376 2009].
377 From the gradients described above, assumed "background" $\Delta^{14}CO_2$ may differ
378 from actual background by as much as 1.5 - 3 ‰ (i.e., ~0.5 - 1 ppm C_{ff}) for large scale,
379 regional studies. In the case of relatively localized campaigns for which up- and down-
380 wind observations within the PBL may be available, the background uncertainty will be

much less and limited primarily by the measurement precision. We note that recent build-out of the observing network in the US, which now includes ~thrice weekly measurements at 9 tall towers (***Fig. 1a***), provides the opportunity to develop a spatially- and temporally- continuous estimate of the $\Delta^{14}CO_2$ field over the US using numerical transport models guided by observations. This, we expect, will substantially reduce both uncertainty and potential bias in C_{ff} arising from uncertainty in "background".

7. Conclusions:

We have demonstrated a scientifically meaningful $\Delta^{14}CO_2$ measurement capability in small (~2L) samples of whole air suitable for implementation within flask sampling networks around the world. The recent measurement repeatability in extraction aliquots of whole air from surveillance cylinders is ≤1.7 ‰ (1σ), and ~1.8 ‰ (1σ) since the inception of our measurement programs in 2003. The latter equates to a fossil fuel CO_2 (C_{ff}) detection capability of 0.9 ppm from measurement uncertainty alone, with an equivalent uncertainty in estimates of the change in CO_2 due to biological uptake and release (C_{bio}). Additional sources of uncertainty in the ^{14}C-based detection of C_{ff} arise from "non-fossil" terms in the isotopic budget, primarily the return flux of $^{14}CO_2$ to the atmosphere from heterotrophic respiration and $^{14}CO_2$ emissions from the nuclear power industry. For the eastern US, we estimate uncertainties from the heterotrophic respiration term of up to ±0.4 ppm C_{ff} in summer and substantially less in winter. Uncertainties associated with variable emissions from the nuclear power sector in the eastern US have a likely maximum of ± 0.1 to ± 1.0 ppm C_{ff}, with a comparable range in the maximum bias in C_{ff} to lower-than-actual values if these emissions are neglected altogether. Uncertainties arising from inadequate observational constraints on "background" $\Delta^{14}CO_2$ are less readily quantified, but may be as large as 0.5 to 1.0 ppm C_{ff} for regional scale (~ 10^5 km^2) monitoring programs [c.f., *Levin et al.*, 2010; *Miller et al.,* 2012]. These uncertainties are likely to be reduced substantially as the increasing number of recent observations

410 are assimilated by transport models in order to represent the time varying $\Delta^{14}CO_2$
411 distribution around the US.
412 As an example of how we apply the measurements, we present estimates of
413 the boundary layer enhancements of C_{ff} and C_{bio} using observations obtained from
414 vertical airborne sampling profiles off of the northeastern US [*Miller et al.*, 2012]. C_{ff}
415 typically ranges from 0-7 ppm, while as much of 60% of the boundary layer CO_2
416 enhancement in winter is from biological respiration. We also present an updated
417 time series of measurements from NOAA GMD's NWR site in CO (at 3475 m asl) in
418 order to characterize recent $\Delta^{14}CO_2$ (and the zonal $\Delta^{14}CO_2$ gradient) in the well-mixed
419 free troposphere over the mid-latitude US.

420

421 **Figure captions:**
422
423 ***Figure 1.*** a) $\Delta^{14}CO_2$ (left panel) and b) the fossil fuel component of total CO_2 (C_{ff},
424 right panel) in the atmosphere near the surface over North America for a week in
425 January of 2006 in the TM5 transport model. The CO_2 and $^{14}CO_2$ budget terms are
426 discussed in the text and at
427 http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/documentation.html. The fossil fuel
428 emissions used in the model are based on the CDIAC US and global totals and the spatial
429 patterns are from the EDGAR inventory. Existing ^{14}C sampling sites for the US are given
430 in a) according to site code (as at http://www.esrl.noaa.gov/gmd/dv/iadv/). Bowties are
431 aircraft sites and triangles are surface or tower sites. NWR (surface flask site) was
432 excluded for clarity, but overlies BAO. Measurements at BAO, LEF and AMT are made in
433 collaboration with investigators at LLNL.
434
435 ***Figure 2.*** $\Delta^{14}CO_2$ measurement values (per mil) in individual extraction aliquots of
436 NWT3 (left panels) or NWT4 (right panels), by order of graphitization and
437 measurement (top panels) or extraction (lower panels). "Manual" (black) and "CRex"
438 (red) refer to results for manual and automated extraction lines, respectively.

439 Uncertainties are given at 1-sigma, and define the longterm repeatability of the
440 measurement, as discussed in the text.
441
442 ***Figure 3*****.** Enhancement or depletion of CO_2 in the planetary boundary layer (~300 m
443 asl) relative to the free troposphere (~4000 m asl) in six years of approximately bi-
444 weekly airborne profiles at CMA and NHA (***Fig. 1a***), separated into biogenic (green)
445 and fossil (red) fractions according to eqs. 2a-c. Black and green lines highlight,
446 respectively, the seasonal variation of raw CO_2 enhancement or depletion in the
447 boundary layer and that attributable to biospheric exchange. The occasional negative
448 values of C_{ff} arise primarily from vertical $\Delta^{14}CO_2$ gradients that are within
449 measurement uncertainties and to a far lesser extent from errors inherent in our
450 simple one-dimensional analytical model.
451
452 ***Figure 4.*** Estimates of C_{corr_resp} for individual ~300 m asl (dark red) and 2000-2400 m asl
453 (light red) samples at CMA, using methods of *Miller et al.* [2012] explained in the text.
454 The monthly means and standard deviations are also shown.
455
456 ***Figure 5.*** Estimates of C_{corr_nuc} for individual ~300 m asl (dark red) and 2000-2400 m asl
457 (light red) samples at CMA, using central, gridded estimates of US nuclear $^{14}CO_2$
458 emissions from *Graven and Gruber* [2011]. The method of calculation is otherwise the
459 same as for C_{corr_resp} (***Fig. 4***). The monthly means and standard deviations are also
460 shown.
461
462 ***Figure 6.*** *a)* $\Delta^{14}CO_2$ measurement values at NWR, CO (3475 m asl, 40.5 °N), for 2003
463 through 2011. Results up to 1/17/06 were presented earlier by *Turnbull et al.* [2007].
464 Measurements for which corresponding CO values exceed a seasonal fit by more than 2
465 sigma were removed to reduce the influence of local pollution events on the record.
466 Remaining results were fitted (black curve) according to the method of *Thoning et al.*
467 [1989]. Individual measurement uncertainties are not shown for clarity, but are

discussed in the text and listed in ***Table 1***. *b)* $\Delta^{14}CO_2$ measurement values for ~4000 m asl samples at NHA (green) and CMA (red), compared to the fitted curve for NWR in *a).* Site locations are given in *Figure 1a.* Note that prior to 8/24/04 at NWR, pilot measurements were made at Rafter Radiocarbon Lab, NZ [*Turnbull et al.*, 2007].

Tables:

Table 1. $\Delta^{14}CO_2$ measurement values at NWR, CO (3475 m asl, 40.5 °N), for 2003 through 2011. *(see separate file)*

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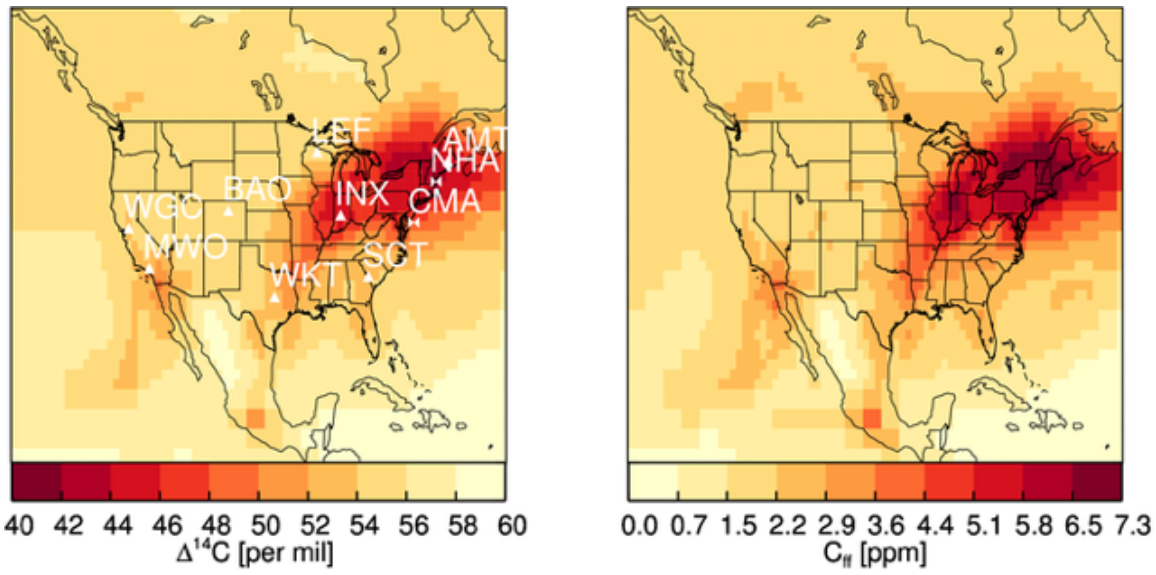

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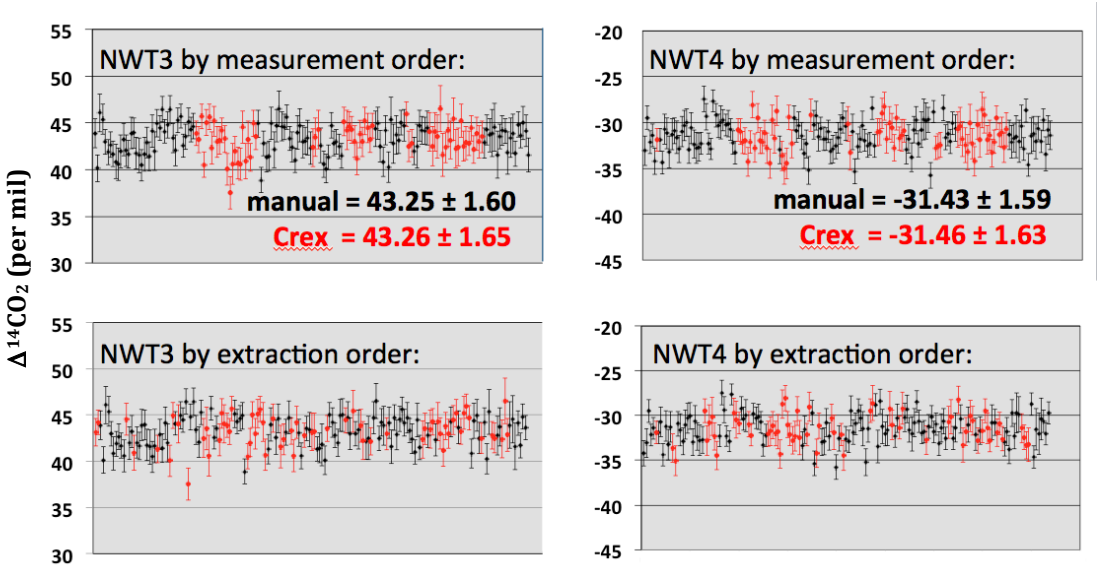
Figure 1.

584



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586
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Figure 2.

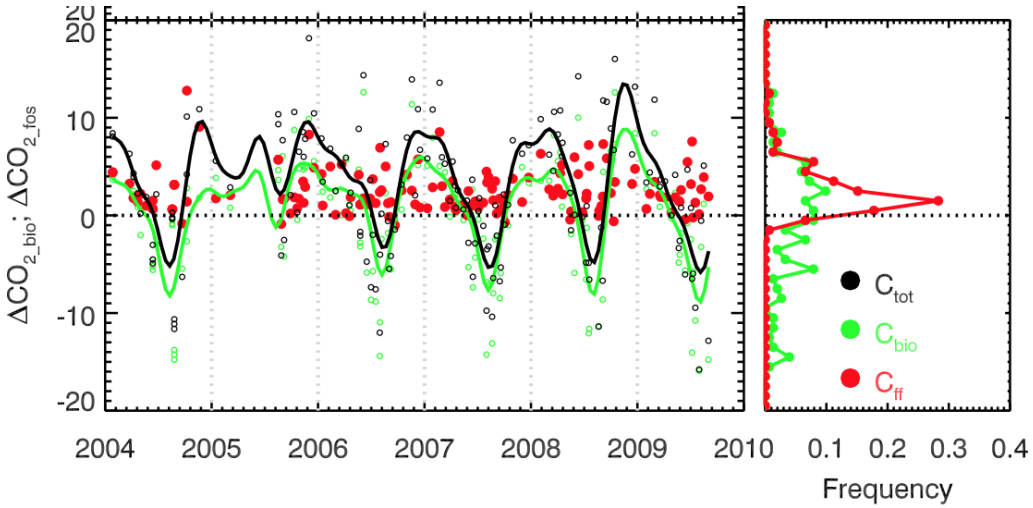
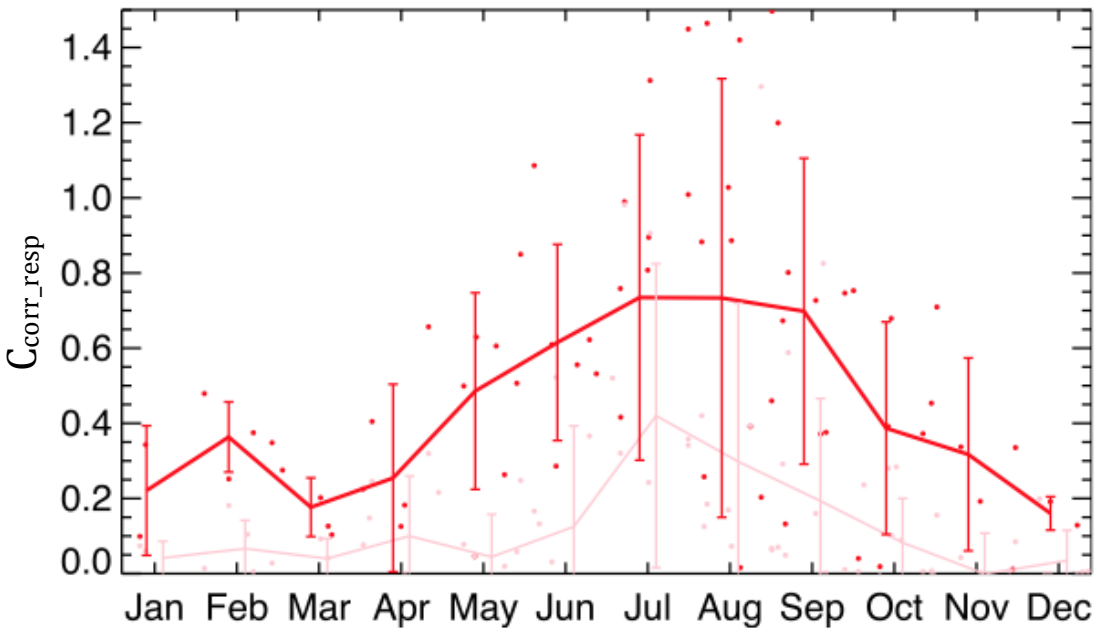
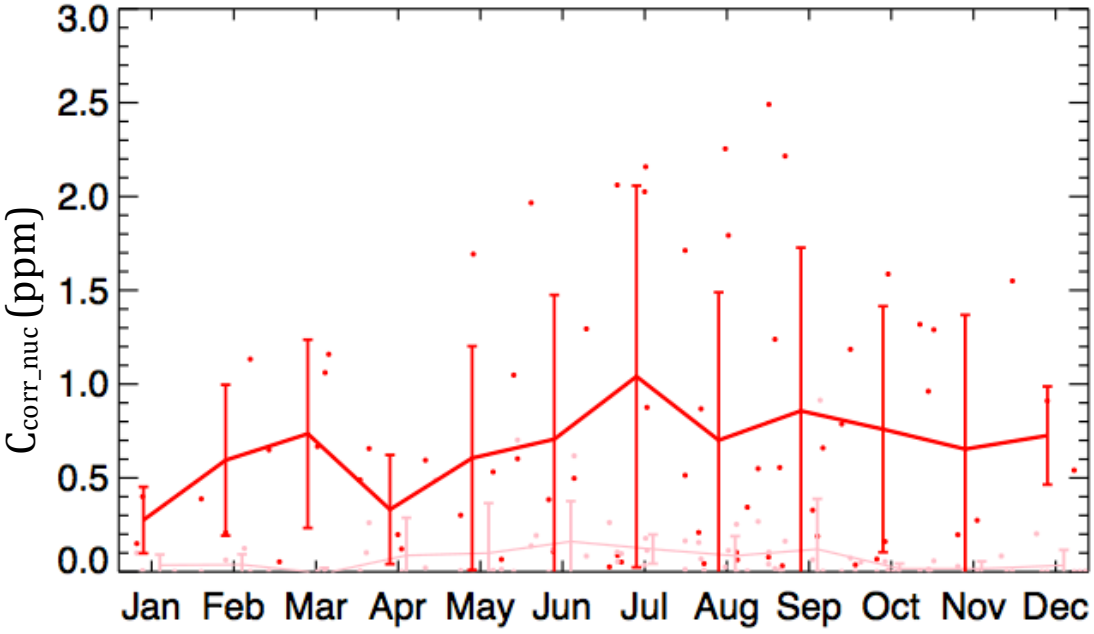

20
10
0
-10
-20
ΔCO2_bio; ΔCO2_fos
2004
2005
2006
2007
2008
2009
2010
0.1
0.2
0.3
0.4
Frequency
Ctot
Cbio
Cff

Figure 3.


1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
C_{corr_resp}
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

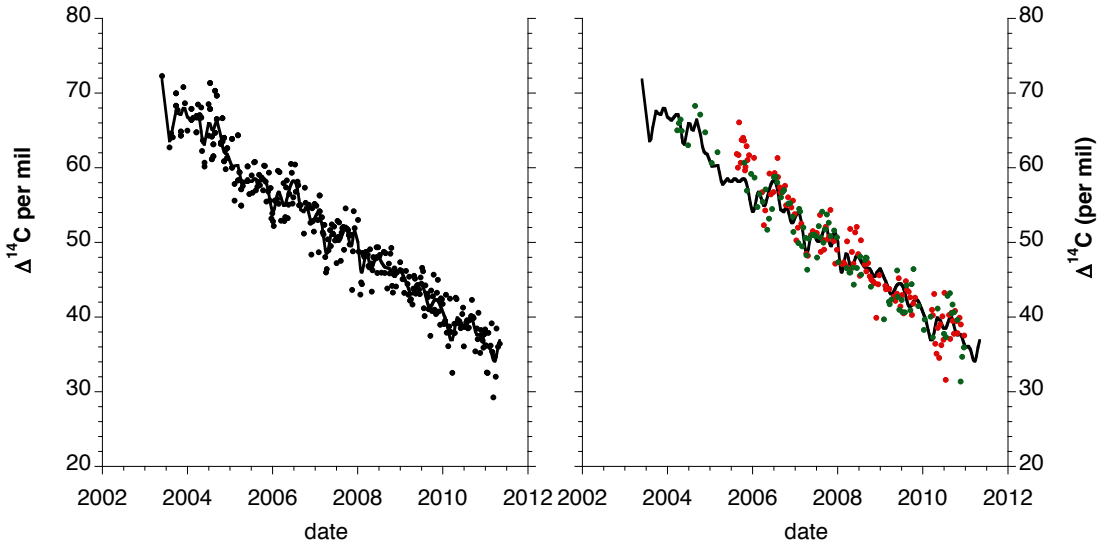
589

Figure 4.


3.0
2.5
2.0
1.5
1.0
0.5
0.0
C_{corr_nuc} (ppm)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

590

Figure 5.



591

592 ***Figure 6.***

yr	mo	day	hr (utc)	dec. date	D14C	D14C_unc
2003	5	27	19	2003.4022	72.29	2.82
2003	5	27	19	2003.4022	72.29	2.82
2003	7	29	16	2003.5745	62.73	2.6
2003	7	29	16	2003.5745	62.73	2.6
2003	8	5	17	2003.5937	65.18	2.6
2003	8	5	17	2003.5937	65.18	2.6
2003	8	12	16	2003.6128	64.61	2.6
2003	8	12	16	2003.6128	64.61	2.6
2003	8	12	16	2003.6128	64.61	2.6
2003	8	12	16	2003.6128	64.61	2.6
2003	8	26	17	2003.6513	64.09	2.6
2003	8	26	17	2003.6513	64.09	2.6
2003	8	26	17	2003.6513	64.09	2.6
2003	8	26	17	2003.6513	64.09	2.6
2003	9	23	20	2003.7284	69.99	2.6
2003	9	23	20	2003.7284	68.32	2.6
2003	9	23	20	2003.7284	69.99	2.6
2003	9	23	20	2003.7284	68.32	2.6
2003	10	7	18	2003.7664	67.76	2.6
2003	10	7	18	2003.7664	67.76	2.6
2003	11	4	21	2003.8436	64.85	2.6
2003	11	4	21	2003.8436	64.33	2.6
2003	11	4	21	2003.8436	64.85	2.6
2003	11	4	21	2003.8436	64.33	2.6
2003	11	25	21	2003.9011	70.82	2.6
2003	11	25	21	2003.9011	70.82	2.6
2003	12	9	19	2003.9393	68.64	2.6
2003	12	9	19	2003.9393	68.64	2.6
2003	12	23	19	2003.9775	66.92	2.6
2003	12	23	19	2003.9775	67.03	2.6
2003	12	23	19	2003.9775	66.92	2.6
2003	12	23	19	2003.9775	67.03	2.6
2003	12	30	19	2003.9967	64.98	2.6
2003	12	30	19	2003.9967	64.98	2.6
2004	2	3	21	2004.0926	67.9	2.6
2004	2	3	21	2004.0926	66.43	2.6
2004	2	3	21	2004.0926	67.9	2.6
2004	2	3	21	2004.0926	66.43	2.6
2004	2	17	20	2004.1307	66.7	2.6
2004	2	17	20	2004.1307	66.7	2.6
2004	3	16	21	2004.2073	66.76	2.6
2004	3	16	21	2004.2073	64.75	2.6

2004	3	16	21	2004.2073	66.76	2.6
2004	3	16	21	2004.2073	64.75	2.6
2004	3	30	17	2004.2451	66.76	2.6
2004	3	30	17	2004.2451	68.48	2.6
2004	3	30	17	2004.2451	66.76	2.6
2004	3	30	17	2004.2451	68.48	2.6
2004	4	13	20	2004.2837	67.07	2.6
2004	4	13	20	2004.2837	67.07	2.6
2004	4	27	20	2004.322	66.74	2.6
2004	4	27	20	2004.322	68.09	2.7
2004	4	27	20	2004.322	66.74	2.6
2004	5	4	20	2004.3412	62.27	2.6
2004	5	4	20	2004.3412	62.27	2.6
2004	5	11	15	2004.3597	63.67	2.6
2004	5	11	15	2004.3597	63.67	2.6
2004	5	25	16	2004.3981	60.69	2.6
2004	5	25	16	2004.3981	60.18	2.6
2004	5	25	16	2004.3981	60.69	2.6
2004	5	25	16	2004.3981	60.18	2.6
2004	6	8	16	2004.4363	66.39	2.6
2004	6	8	16	2004.4363	62.43	2.6
2004	6	8	16	2004.4363	66.39	2.6
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2004	6	29	18	2004.4939	64.32	2.6
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2004	7	27	15	2004.5701	61.14	2.6
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2004	8	10	17	2004.6086	61.55	2.6
2004	8	10	17	2004.6086	63.15	2.6
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2004	8	24	19	2004.647	64.69	2.6
2004	8	24	19	2004.647	70.32	2.6
2004	8	24	19	2004.647	64.69	2.6

2004	9	7	19	2004.6852	69.67	2.7
2004	9	7	19	2004.6852	66.52	1.8
2004	9	7	19	2004.6852	69.67	2.7
2004	9	7	19	2004.6852	66.52	1.8
2004	9	28	20	2004.7428	62.55	2.7
2004	9	28	20	2004.7428	63.13	1.8
2004	9	28	20	2004.7428	62.55	2.7
2004	9	28	20	2004.7428	63.13	1.8
2004	10	12	17	2004.7807	63.38	2.74
2004	10	12	17	2004.7807	63.22	1.8
2004	10	12	17	2004.7807	63.38	2.74
2004	10	12	17	2004.7807	63.22	1.8
2004	10	26	19	2004.8192	63.54	2.7
2004	10	26	19	2004.8192	66.66	1.8
2004	10	26	19	2004.8192	63.54	2.7
2004	10	26	19	2004.8192	66.66	1.8
2004	11	9	22	2004.8577	64.03	2.7
2004	11	9	22	2004.8577	60.89	1.8
2004	11	9	22	2004.8577	64.03	2.7
2004	11	9	22	2004.8577	60.89	1.8
2004	11	23	22	2004.8959	62.41	2.7
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2004	11	23	22	2004.8959	62.41	2.7
2004	11	23	22	2004.8959	59.82	1.8
2004	12	7	22	2004.9343	61.34	2.9
2004	12	7	22	2004.9343	62.63	1.8
2004	12	7	22	2004.9343	61.34	2.9
2004	12	7	22	2004.9343	62.63	1.8
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2005	1	17	21	2005.0463	63.88	1.8
2005	1	17	21	2005.0463	60.04	2.73
2005	1	17	21	2005.0463	63.88	1.8
2005	2	8	22	2005.1066	57.84	2.7
2005	2	8	22	2005.1066	55.61	1.8
2005	2	8	22	2005.1066	57.84	2.7
2005	2	8	22	2005.1066	55.61	1.8
2005	3	8	21	2005.1833	58.32	2.7
2005	3	8	21	2005.1833	64.36	1.8
2005	3	8	21	2005.1833	58.32	2.7
2005	3	8	21	2005.1833	64.36	1.8
2005	3	22	22	2005.2217	58.35	2.7
2005	3	22	22	2005.2217	59.4	1.8
2005	3	22	22	2005.2217	58.35	2.7

2005	3	22	22	2005.2217	59.4	1.8
2005	4	5	19	2005.2598	54.95	2.7
2005	4	5	19	2005.2598	57.13	1.8
2005	4	5	19	2005.2598	54.95	2.7
2005	4	5	19	2005.2598	57.13	1.8
2005	4	19	20	2005.2983	57.57	2.7
2005	4	19	20	2005.2983	58.06	1.8
2005	4	19	20	2005.2983	57.57	2.7
2005	4	19	20	2005.2983	58.06	1.8
2005	5	31	18	2005.413	60.21	2.7
2005	5	31	18	2005.413	58.24	1.8
2005	5	31	18	2005.413	60.21	2.7
2005	5	31	18	2005.413	58.24	1.8
2005	6	14	18	2005.4515	53.6	2.7
2005	6	14	18	2005.4515	59.34	1.8
2005	6	14	18	2005.4515	53.6	2.7
2005	6	14	18	2005.4515	59.34	1.8
2005	6	14	18	2005.4515	53.6	2.7
2005	6	14	18	2005.4515	59.34	1.8
2005	7	5	14	2005.5085	58.97	1.8
2005	7	5	14	2005.5085	58.97	1.8
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2005	7	12	17	2005.528	56.67	1.8
2005	7	19	15	2005.5469	60.2	1.8
2005	7	19	15	2005.5469	60.2	1.8
2005	7	26	15	2005.5661	60.73	1.8
2005	7	26	15	2005.5661	60.73	1.8
2005	8	2	14	2005.5852	60.8	1.8
2005	8	2	14	2005.5852	60.8	1.8
2005	8	9	15	2005.6045	56.98	1.8
2005	8	9	15	2005.6045	56.98	1.8
2005	8	16	17	2005.6239	65.12	2.18
2005	8	16	17	2005.6239	65.12	2.18
2005	8	23	15	2005.6428	57.48	1.8
2005	8	23	15	2005.6428	57.48	1.8
2005	8	30	16	2005.6621	58.08	1.8
2005	8	30	16	2005.6621	58.08	1.8
2005	9	6	16	2005.6813	59.07	1.8
2005	9	6	16	2005.6813	59.07	1.8
2005	9	13	16	2005.7005	57.67	1.8
2005	9	13	16	2005.7005	57.67	1.8
2005	9	27	17	2005.739	58.72	1.8
2005	9	27	17	2005.739	58.72	1.8

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2010	1	12	23	2010.0328	39.1	1.8
2010	1	12	23	2010.0328	39.1	1.8
2010	1	19	22	2010.0518	39.53	1.8
2010	1	19	22	2010.0518	39.53	1.8
2010	1	26	20	2010.0708	43	1.8
2010	1	26	20	2010.0708	43	1.8
2010	2	2	22	2010.0903	37.91	1.8
2010	2	2	22	2010.0903	37.91	1.8
2010	2	16	22	2010.1286	36.12	1.8
2010	2	16	22	2010.1286	36.12	1.8
2010	2	23	20	2010.1476	38.77	1.8
2010	2	23	20	2010.1476	38.77	1.8
2010	3	2	21	2010.1668	42.47	1.8
2010	3	2	21	2010.1668	42.47	1.8
2010	3	9	22	2010.1861	38.04	1.8
2010	3	9	22	2010.1861	38.04	1.8
2010	3	23	20	2010.2242	32.55	1.8

2010	3	23	20	2010.2242	32.55	1.8
2010	4	8	20	2010.2681	39.12	1.8
2010	4	8	20	2010.2681	39.12	1.8
2010	4	13	19	2010.2817	37.89	1.83
2010	4	13	19	2010.2817	37.89	1.83
2010	4	20	20	2010.301	42.61	1.8
2010	4	20	20	2010.301	42.61	1.8
2010	4	27	21	2010.3202	42.43	1.8
2010	4	27	21	2010.3202	42.43	1.8
2010	5	4	21	2010.3394	39.21	1.8
2010	5	4	21	2010.3394	39.21	1.8
2010	6	1	19	2010.4159	38.54	1.8
2010	6	1	19	2010.4159	38.54	1.8
2010	6	8	19	2010.4351	38.88	1.8
2010	6	8	19	2010.4351	38.88	1.8
2010	6	15	17	2010.454	40.43	1.8
2010	6	15	17	2010.454	40.43	1.8
2010	6	22	17	2010.4732	41.28	1.8
2010	6	22	17	2010.4732	41.28	1.8
2010	6	29	16	2010.4923	36.1	1.8
2010	6	29	16	2010.4923	36.1	1.8
2010	7	13	17	2010.5307	38.8	1.8
2010	7	13	17	2010.5307	38.8	1.8
2010	7	20	17	2010.5499	39.32	1.8
2010	7	20	17	2010.5499	39.32	1.8
2010	7	27	16	2010.569	39.58	1.8
2010	7	27	16	2010.569	39.58	1.8
2010	8	3	19	2010.5885	38.5	1.8
2010	8	3	19	2010.5885	38.5	1.8
2010	8	10	20	2010.6078	38.6	1.8
2010	8	10	20	2010.6078	38.6	1.8
2010	8	17	16	2010.6266	39.59	1.8
2010	8	17	16	2010.6266	39.59	1.8
2010	8	31	19	2010.6652	40.15	1.8
2010	8	31	19	2010.6652	40.15	1.8
2010	9	7	18	2010.6842	42.31	1.8
2010	9	7	18	2010.6842	42.31	1.8
2010	9	14	19	2010.7036	40.06	1.8
2010	9	14	19	2010.7036	40.06	1.8
2010	9	21	20	2010.7229	39.89	1.8
2010	9	21	20	2010.7229	39.89	1.8
2010	10	5	21	2010.7613	35.43	1.8
2010	10	5	21	2010.7613	35.43	1.8

2010	10	12	17	2010.7801	35.63	1.8
2010	10	12	17	2010.7801	35.63	1.8
2010	10	19	17	2010.7992	38.48	1.8
2010	10	19	17	2010.7992	38.48	1.8
2010	10	26	20	2010.8188	37.05	1.8
2010	10	26	20	2010.8188	37.05	1.8
2010	11	2	17	2010.8376	40.59	1.8
2010	11	2	17	2010.8376	40.59	1.8
2010	11	16	21	2010.8765	35.22	1.8
2010	11	16	21	2010.8765	35.22	1.8
2010	11	23	22	2010.8957	38.28	1.8
2010	11	23	22	2010.8957	38.28	1.8
2010	11	30	21	2010.9148	37.28	1.84
2010	11	30	21	2010.9148	37.28	1.84
2010	12	7	22	2010.9341	36.87	1.8
2010	12	7	22	2010.9341	36.87	1.8
2010	12	14	22	2010.9533	35.91	1.8
2010	12	14	22	2010.9533	35.91	1.8
2010	12	21	17	2010.9719	37.72	1.8
2010	12	21	17	2010.9719	37.72	1.8
2010	12	28	18	2010.9912	38.37	1.8
2010	12	28	18	2010.9912	38.37	1.8
2011	1	4	19	2011.0105	36.24	1.81
2011	1	4	19	2011.0105	36.24	1.81
2011	1	11	22	2011.0299	32.6	1.8
2011	1	11	22	2011.0299	32.6	1.8
2011	1	18	20	2011.049	32.53	1.8
2011	1	18	20	2011.049	32.53	1.8
2011	1	25	17	2011.0678	37.9	1.8
2011	1	25	17	2011.0678	37.9	1.8
2011	2	14	18	2011.1226	36.21	1.8
2011	2	14	18	2011.1226	36.21	1.8
2011	2	22	21	2011.1449	39.17	1.8
2011	2	22	21	2011.1449	39.17	1.8
2011	2	28	22	2011.1614	35.37	1.8
2011	2	28	22	2011.1614	35.37	1.8
2011	3	9	20	2011.1859	29.26	1.81
2011	3	9	20	2011.1859	29.26	1.81
2011	3	15	18	2011.2021	37.22	1.8
2011	3	15	18	2011.2021	37.22	1.8
2011	3	22	20	2011.2215	34.76	1.8
2011	3	22	20	2011.2215	34.76	1.8
2011	3	29	18	2011.2405	32.03	1.8

2011	3	29	18	2011.2405	32.03	1.8
2011	4	5	18	2011.2596	38.49	1.8
2011	4	5	18	2011.2596	38.49	1.8
2011	4	19	17	2011.2979	36.03	1.8
2011	4	19	17	2011.2979	36.03	1.8
2011	5	3	22	2011.3368	36.44	1.8
2011	5	3	22	2011.3368	36.44	1.8