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$\delta^{13}C$ of CFC-12
$\delta^{13}C$ of CFC-12

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Heptafluoropropane in atmosphere
CO budget

Modeling the recent anthropogenic impact on the atmosphere from polar firn measurements

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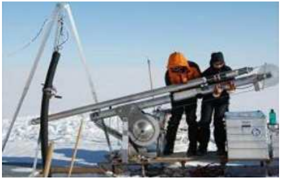
Motivation

Trace gas measurements in interstitial air from polar firn

- allow to reconstruct their atmospheric concentration time trends over the last 50 to 100 years
- provides a unique way to reconstruct the recent anthropogenic impact on atmospheric composition

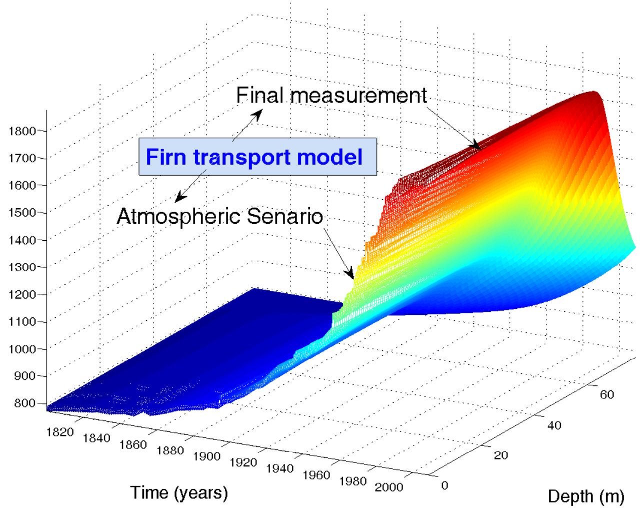


Converting depth-concentration profiles in firn into atmospheric concentration histories requires models of trace gas transport in firn

Background : previous (and first) version of LGGE firn models : Rommelaere et al., Ph.D. 1995, JGR, 1997

Direct model

I.e. CH_4 transport at NEEM (Greenland)



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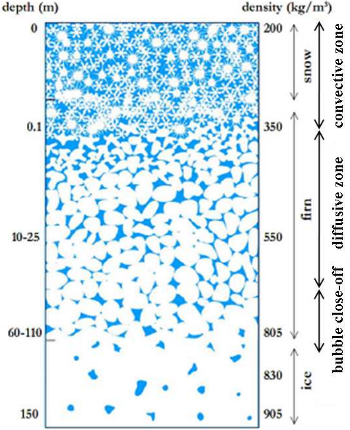
Poromechanics : three interconnected networks

Ice lattice, gas connected to the surface (open pores) and gas trapped in bubbles (closed pores) :

$$\frac{\partial[\rho_{ice}(1-\epsilon)]}{\partial t} + \nabla[\rho_{ice}(1-\epsilon)\vec{v}] = 0$$

$$\frac{\partial[\rho^o_{gas} f]}{\partial t} + \nabla[\rho^o_{gas} f(\vec{v} + \vec{w}_{gas})] = -\vec{r}^{o\to c}$$

$$\frac{\partial[\rho^c_{gas}(\epsilon - f)]}{\partial t} + \nabla[\rho^c_{gas}(\epsilon - f)\vec{v}] = \vec{r}^{o\to c}$$



Scheme adapted from [Sowers et al.'92, Lourantou'08].

Trace gas conservation in open pores [Rommelaere & al.'97, Witrant & al.'11]

- Flux driven by advection with air and firn sinking
- Flux driven by mol. diff. due to concentration gradients
- Flux driven by external forces : gravity included with Darcy-like flux
- Sink = particles trapped in bubbles & radioactive decay
- Boundary input : surface concentration
- Results in transport PDE :

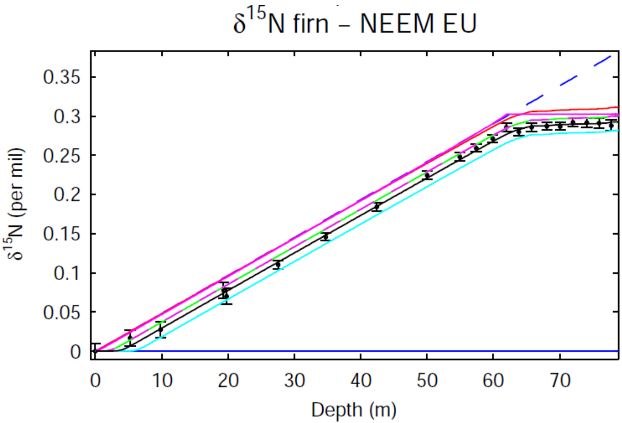
$$\frac{\partial}{\partial t}[\rho_\alpha^o f] + \frac{\partial}{\partial z}[\rho_\alpha^o f(v + w_{air})] + \rho_\alpha^o(\tau + \lambda) - \frac{\partial}{\partial z}\left[\mathbf{D}_\alpha\left(\frac{\partial \rho_\alpha^o}{\partial z} - \rho_\alpha^o \frac{\partial \rho_{air}/\partial z}{\rho_{air}} + \mathcal{A}_{ss}\right)\right] = 0$$

$$\rho_\alpha^o(0,t) = \rho_\alpha^{atm}(t), \qquad \frac{RT}{M_f}\frac{\partial \rho_\alpha^o}{\partial z}(z_f) - \rho_\alpha^o(z_f) = 0$$

with $\mathcal{A}_{ss}$ such that $\partial[\rho_{\alpha,ss}^o f]/\partial t = 0$ at steady state, i.e.

$$\mathcal{A}_{ss} = -\frac{\rho_{\alpha,ss}^o f}{D_\alpha}(w_\alpha - w_{air}) - \left(\frac{\partial \rho_{\alpha,ss}^o}{\partial z} - \rho_{\alpha,ss}^o \frac{\partial \rho_{air}/\partial z}{\rho_{air}}\right)$$

Validation on isotopic indicators : $\delta^{15}N$ ($\delta^{40}Ar$, $\delta^{86}Kr$)



Fick only (blue '—'), QSS (exact in blue '- - -' and gas speed set by air speed in red), QSS with forced LIZ (pink '—'), QSS with $z_{conv} = 4$ m : hydrostatic $\rho^o_{\alpha,ss}$ (green), + max D set by the one in free air + gas-indep term (pink '- - -'), + $z_{conv} = z_{eddy}$ (turquoise), Ref case (black) : simplified QSS with $z_{conv} = 4$ m and a max mol. diffu. corrected with the porosity

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Problem formulation

- Least squares minimization (single gas) :

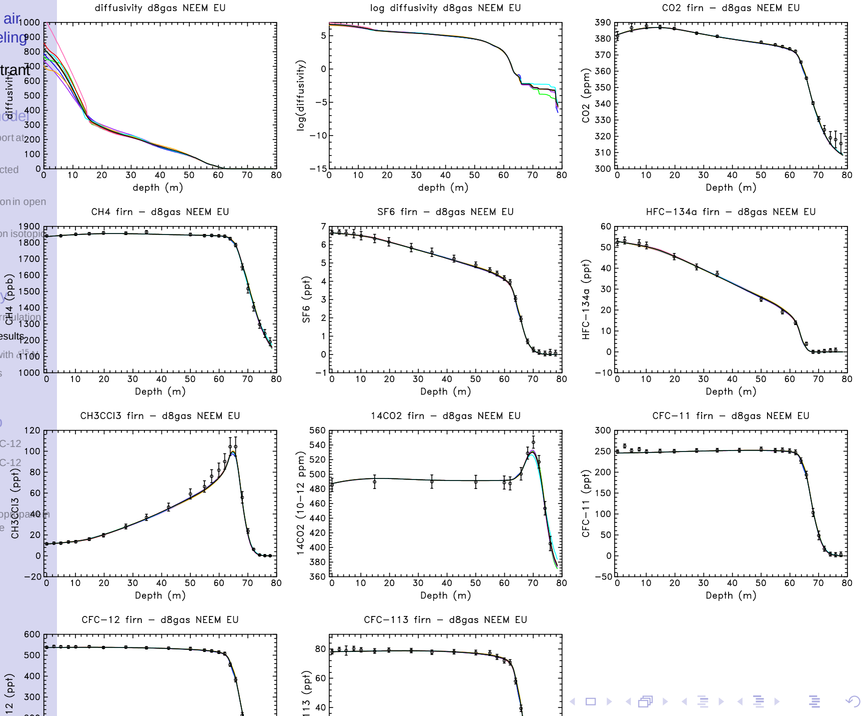
$$D_\alpha^* = \arg\min_{D_\alpha} \frac{1}{z_f} \int_0^{z_f} \frac{1}{\sigma_\alpha^2} \left(m_\alpha - \frac{\rho_\alpha^o(D_\alpha)}{\rho_{air}^o} \right)^2 \delta_\alpha dz$$

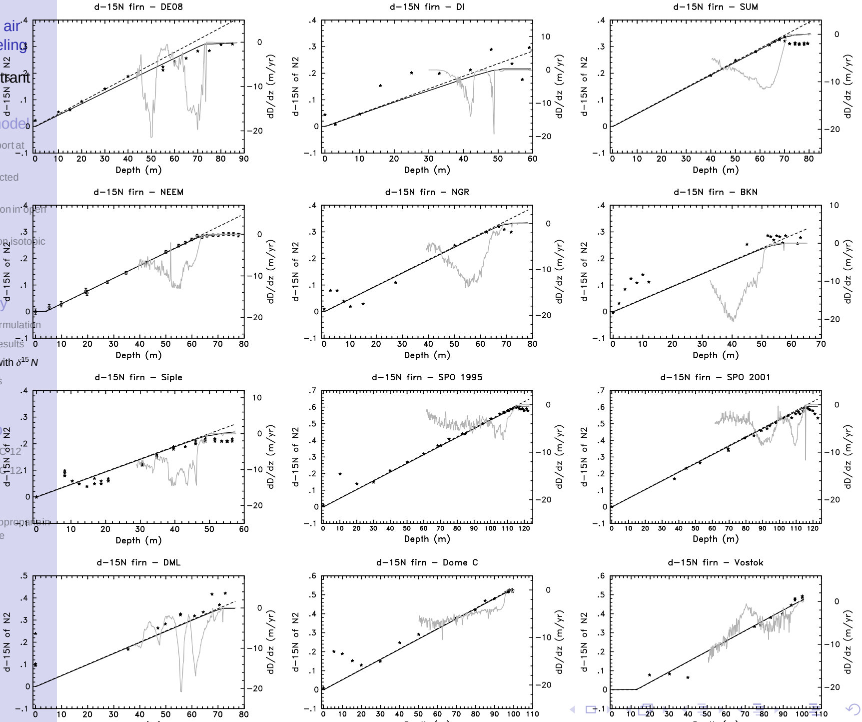
  with the constraints $D(z) > 0$ and $dD/dz < 0$

- For N gas :

$$D_{CO_2}^* = \arg\min_{D_{CO_2}} \sum_{i=1}^{N} \frac{1}{z_f} \int_0^{z_f} \frac{1}{\sigma_{\alpha_i}^2} \left(m_{\alpha_i} - \frac{\rho_{\alpha_i}^o(D_{CO_2})}{\rho_{air}^o} \right)^2 \delta_{\alpha_i} dz$$

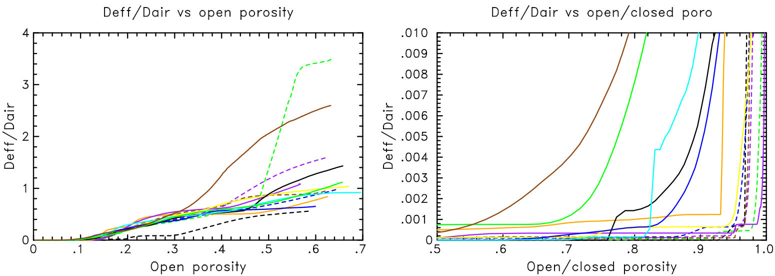
- Nonlinear optimization problem (at least with implicit schemes)


diffusivity d8gas NEEM EU
diffusivity
depth (m)
log diffusivity d8gas NEEM EU
log(diffusivity)
depth (m)
CO2 firn – d8gas NEEM EU
CO2 (ppm)
Depth (m)
CH4 firn – d8gas NEEM EU
CH4 (ppb)
Depth (m)
SF6 firn – d8gas NEEM EU
SF6 (ppt)
Depth (m)
HFC–134a firn – d8gas NEEM EU
HFC–134a (ppt)
Depth (m)
CH3CCl3 firn – d8gas NEEM EU
CH3CCl3 (ppt)
Depth (m)
14CO2 firn – d8gas NEEM EU
14CO2 (10–12 ppm)
Depth (m)
CFC–11 firn – d8gas NEEM EU
CFC–11 (ppt)
Depth (m)
CFC–12 firn – d8gas NEEM EU
12 (ppt)
CFC–113 firn – d8gas NEEM EU
113 (ppt)



d−15N firn − DE08
d−15N firn − DI
d−15N firn − SUM
d−15N firn − NEEM
d−15N firn − NGR
d−15N firn − BKN
d−15N firn − Siple
d−15N firn − SPO 1995
d−15N firn − SPO 2001
d−15N firn − DML
d−15N firn − Dome C
d−15N firn − Vostok
d−15N of N2
dD/dz (m/yr)
Depth (m)

Result = Diffusivities at 11 sites (13 holes) [ACPD'11]



Arctic (dashed) : Devon Island (black), Summit (blue), NEEM-EU (purple) and NEEM-US (brown), North GRIP (green).
Antarctic (continuous) : DE08 (orange), Berkner (purple), Siple (yellow), South Pole 1995 (dark blue), South Pole 2001 (light blue), Dronning Maud Land (black), Dome C (green) and Vostok (brown)

- Low diffusivity at Devon Island due to melt layers
- High diffusivity in upper firn related to convection
- Very consistent diff. at intermediate depths (0.1-0.3)
- High diff. in deep firn at Vostok and Dome C (low accu. and cold), consistent with very young ages and no plateau in $\delta^{15}N$

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Inverse scenario model

A "deconvolution" approach [Rommelaere et al., JGR, 1997]

- Green function = impulse response of the firn ⇒ age probabilities

$$\rho_{firn}(z, t_f) = G(z, t) * \rho_{atm}(t) \qquad \text{convolution}$$

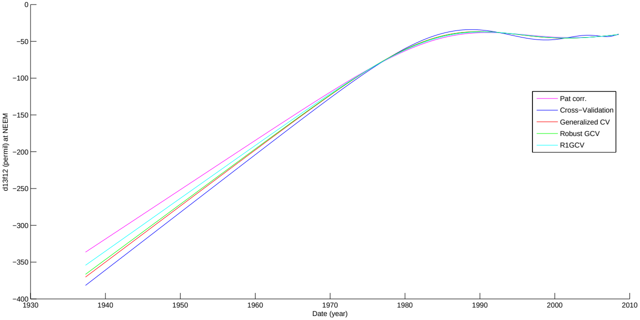
- Deconvolution :

$$\begin{aligned} \epsilon(z) &= G(z, t)\rho_{atm}(t) - \rho_{firn}(z, t_f) \\ \rho^*_{atm}(t) &= arg\min_{\rho_{atm}} \left[\epsilon^T (diag\{1/\sigma^2_{mes}(z)\})\epsilon + \kappa^2 \rho^T_{atm} R \rho_{atm}\right] \end{aligned}$$

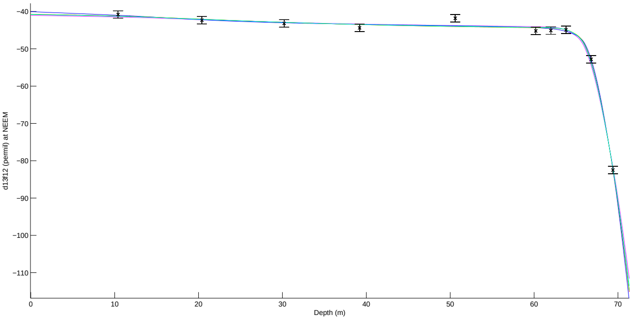
- Under-constrained pb ⇒ add extra information with rugosity characteristic matrix $R > 0$ (i.e. d^2/dt^2) + κ.
- 2 parameters largely control model behavior : κ (rugosity factor) and $\sigma^2_{mes}(z)$

⇒ Extension to multi-site and isotopic ratios (time-varying parameters, robust cross-validation)

Inverse scenario for δ^{13}C of CFC-12 at NEEM EU 2008 [Zuiderweg et al. 2012]

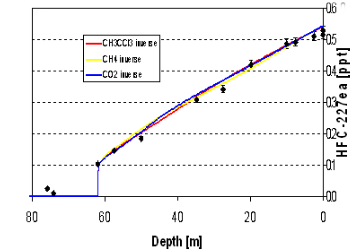


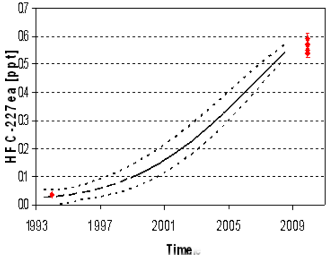
Resulting firn concentrations



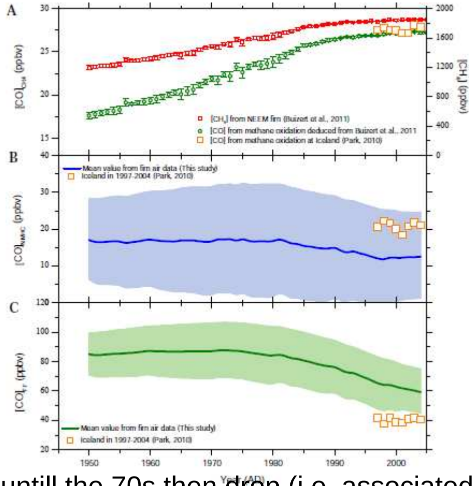
Accelerating growth of HFC-227ea in the atmosphere [Laube *et. al*'10]

- HFC-227ea = substitute for ozone depleting compounds
- Firn air samples collected in Greenland used to reconstruct a history of atmospheric abundance from 2000 to 2007
- Acceleration in growth rate confirmed by upper tropospheric air samples in 2009
- Stratospheric lifetime of 370 years calculated with samples from high altitude aircraft and balloons





The isotopic record of Northern Hemisphere atmospheric carbon monoxide since 1950, implications for the CO budget [Wang *et. al*'12]



⇒ Increase untill the 70s then drop (i.e. associated with fossil fuel : catalytic converters and diesel engines)

Other results

- Atmospheric impacts and ice core imprints of a methane pulse from clathrates [Bock *et. al*'12]
- Extreme ^{13}C depletion of CCl2F2 in firn air samples from NEEM, Greenland [Zuiderweg *et. al*'12]
- Emissions halted of the potent greenhouse gas SF5CF3 [Sturges *et. al*'12]
- Distributions, long term trends and emissions of four perfluorocarbons in remote parts of the atmosphere and firn air [Laube *et. al*'12]
- Reconstruction of the carbon isotopic composition of methane over the last 50 yr based on firn air measurements at 11 polar sites [Sapart *et. al*'12]
- Natural and anthropogenic variations in methane sources over the last 2 millennia [Sapart *et. al*'12]