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Supplement of

Novel Keeling-plot-based methods to estimate the isotopic composition of ambient water vapor

Yusen Yuan et al.

Correspondence to: Lixin Wang (wang.iupui@gmail.com) and Taisheng Du (dutaisheng@cau.edu.cn)

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Proposition. In the traditional linear Keeling plot system, denote $\delta_a = f(t)$, $\delta_v = g(t)$, $\delta_{ET} = h(t)$ and $C_a = I(t) > 0$ as continuous functions of time. And for two definite moments t_1 and t_2 ($t_1 < t_2$), $\delta_{a_1} \neq \delta_{a_2} \neq \delta_{v_1} \neq \delta_{v_2} \neq \delta_{ET_1} \neq \delta_{ET_2}$. The slopes of corresponding keeling plot curve are $k_1 = C_{a_1}(\delta_{a_1} - \delta_{ET_1})$ and $k_2 = C_{a_2}(\delta_{a_2} - \delta_{ET_2})$, respectively. Then we have that when $k_1 k_2 < 0$, there exists $[t_1', t_2'] \subset [t_1, t_2]$, such that $[\min(f(t_1'), f(t_2')), \max(f(t_1'), f(t_2'))] \subset [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$.

Remark: To make a proof of the proposition, classical Intermediate Value Theorem (IVT) was used. It states that if f is a continuous function from the interval $I = [a, b]$ to real number (R). Then *Version I*. if u is a number between $f(a)$ and $f(b)$, there is c in (a, b) such that $f(c) = u$. *Version II*. the image set $f(I)$ is also an interval, and it contains $[\min(f(a), f(b)), \max(f(a), f(b))]$. While in this study, IVT was able to be explained as follows: if f is a continuous function from the interval $I = [t_1, t_2]$ to R with $\min[f(t_1), f(t_2)] < \delta_v$ and $\max[f(t_1), f(t_2)] > \delta_v$, then *Version I* implies that there is $t' \in (t_1, t_2)$ such that $f(t') = \delta_v$. And *Version II* implies that the image set $f(I)$ is also an interval, and it contains $[\min(f(t_1), f(t_2)), \max(f(t_1), f(t_2))]$.

Proof. Since $k_1 k_2 < 0$, we have $\delta_{a_1} < \delta_{v_1}$ and $\delta_{a_2} > \delta_{v_2}$, or $\delta_{a_1} > \delta_{v_1}$ and $\delta_{a_2} < \delta_{v_2}$. As a result, the cases $\delta_{a_1} < \delta_{v_1} < \delta_{a_2} < \delta_{v_2}$, $\delta_{v_1} < \delta_{a_1} < \delta_{v_2} < \delta_{a_2}$, $\delta_{v_2} < \delta_{a_2} < \delta_{v_1} < \delta_{a_1}$, $\delta_{a_2} < \delta_{v_2} < \delta_{a_1} < \delta_{v_1}$ and $[\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})] \cap [\min(\delta_{a_1}, \delta_{a_2}), \max(\delta_{a_1}, \delta_{a_2})] = \emptyset$ do not meet the precondition $k_1 k_2 < 0$. There are only four cases below. We will prove the proposition in each of the four cases.

Case 1: $[\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})] \subset [\min(\delta_{a_1}, \delta_{a_2}), \max(\delta_{a_1}, \delta_{a_2})]$ (**Fig. 1 a**).

According to IVT *Version I*, there exists $t_1' \in [t_1, t_2]$, such that $f(t_1') = \delta_{v_1}$; similarly, there exists $t_2' \in [t_1, t_2]$, such that $f(t_2') = \delta_{v_2}$. Based on IVT *Version II*, there exists $[t_1', t_2'] \subset [t_1, t_2]$, such that $[\min(f(t_1'), f(t_2')), \max(f(t_1'), f(t_2'))] = [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$.

Case 2: $[\min(\delta_{a_1}, \delta_{a_2}), \max(\delta_{a_1}, \delta_{a_2})] \subset [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$ (**Fig. 1 b**).

According to IVT *Version I*, there exists $t_1' \in [t_1, t_2]$, such that $f(t_1') = \delta_{a_1}$; similarly, there exists $t_2' \in [t_1, t_2]$, such that $f(t_2') = \delta_{a_2}$. Based on IVT *Version II*, there exists $[t_1', t_2'] \subset [t_1, t_2]$, such that $[\min(f(t_1'), f(t_2')), \max(f(t_1'), f(t_2'))] = [\min(\delta_{a_1}, \delta_{a_2}), \max(\delta_{a_1}, \delta_{a_2})] \subset [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$.

Case 3: $\delta_{v_2} < \delta_{a_1} < \delta_{v_1} < \delta_{a_2}$, or $\delta_{a_2} < \delta_{v_1} < \delta_{a_1} < \delta_{v_2}$ (**Fig. 1 c and Fig. 1 d**).

According to IVT Version I, there exists $t_2' \in [t_1, t_2]$, such that $f(t_2') = \delta_{v_1}$. Given case (2), when $[\min(\delta_{a_1}, \delta_{v_1}), \max(\delta_{a_1}, \delta_{v_1})] \subset [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$, there exists $[t_1', t_2'] \subset [t_1, t_2]$, such that $[\min(f(t_1'), f(t_2')), \max(f(t_1'), f(t_2'))] \subset [\min(\delta_{a_1}, \delta_{v_1}), \max(\delta_{a_1}, \delta_{v_1})] \subset [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$.

Case 4: $\delta_{v_1} < \delta_{a_2} < \delta_{v_2} < \delta_{a_1}$, or $\delta_{a_1} < \delta_{v_2} < \delta_{a_2} < \delta_{v_1}$ (**Fig. 1 e and Fig.1 f**).

According to IVT Version I, there exists $t_1' \in [t_1, t_2]$, such that $f(t_1') = \delta_{v_2}$. Based on case (2), when $[\min(\delta_{a_2}, \delta_{v_2}), \max(\delta_{a_2}, \delta_{v_2})] \subset [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$, there exists $[t_1', t_2'] \subset [t_1, t_2]$, such that $[\min(f(t_1'), f(t_2')), \max(f(t_1'), f(t_2'))] \subset [\min(\delta_{a_2}, \delta_{v_2}), \max(\delta_{a_2}, \delta_{v_2})] \subset [\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$.

Thus the proposition is true for all four possible scenarios, which make the estimation of δ_a theoretically feasible when $k_1 k_2 < 0$ and δ_{v_1} and δ_{v_2} adequately close. Actual δ_a between t_1 and t_2 can be ensured in the interval $[\min(\delta_{v_1}, \delta_{v_2}), \max(\delta_{v_1}, \delta_{v_2})]$. To

simplify the result, actual δ_a between t_1 and t_2 can be approximately regarded as what Eq. (7) reveals.